Electron Correlation in Ferromagnetism

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The effect of electron correlation on the ferromagnetism of a transition metal is investigated by taking an approximate model Hamiltonian which takes into account the effect of interatomic interactions. An approximate solution of the correlation problem for this model is obtained by using the Green-function method. The condition for ferromagnetism for this solution is discussed.

I. INTRODUCTION

In recent years, the awareness of the role of correlation among electrons in the magnetic properties of metals has motivated several treatments of correlation effects in transition metals.[1] The theory of electrons with strong correlations in a narrow s band has been developed by Kanamori,[2] by Gutzwiller,[3] and by Hubbard[4,5] using three different methods. Kanamori uses the two-body scattering operators, Gutzwiller utilizes the correlated wave functions, and Hubbard employs the one-particle Green functions. These authors examined the magnetic stability of the system. Several other authors[5–9] have also discussed the stability of ferromagnetic state by using the Hubbard model. In all these treatments, it is assumed that only intra-atomic interaction between the electrons is appreciable, and interatomic interactions can be neglected. In spite of various approaches, models and approximations proposed to explain ferromagnetism in metals, our understanding of the origin of ferromagnetism is still meager. Therefore, it still has some meaning to study a simple model in detail and to examine whether it can be ferromagnetic or not.

In this paper, we study a model for a narrow s band of electrons described by the Hamiltonian

$$H=\sum_{ij} T_{ij}c_{i\sigma}^{\dagger}c_{j\sigma}+\frac{I}{2}\sum_{i\sigma} n_{i\sigma}n_{i-\sigma}+\tfrac{1}{2}\sum_{\substack{ij\\ \sigma\sigma'}} Q_{ij}n_{i\sigma}n_{j\sigma'}. \quad (1)$$

The derivation of the Hamiltonian, various approximations implied in it, and the meaning of symbols are discussed in the Appendix. This Hamiltonian differs from that of Hubbard[4] by the inclusion of third term on the right-hand side of Eq. (1). This term represents the interatomic correlation of electrons at two different sites.

Our approach to the correlation problem is based on the Green-function method discussed by Zubarev.[10] In Sec. II, an approximate theory for correlation effects in electrons is developed. Here, we have evaluated one-electron Green function for the system described by the Hamiltonian (1). The higher-order Green functions appearing in the equations of motion of Green function are decoupled within the Hartree-Fock approximation and within the approximations similar to that of Hubbard.[4] We also find the approximate solution of correlation problem in zero bandwidth limit. The same problem is then discussed for finite bandwidth case. In Sec. III, the occurrence of ferromagnetism for square density of states is discussed. Section IV summarizes our findings.

II. SOME APPROXIMATE SOLUTIONS

The double-time temperature-dependent retarded (+) and advanced (−) Green functions involving two operators A and B are defined by

$$\langle\langle A(t);B(t')\rangle\rangle^{(\pm)}=\mp i\theta\{\pm(t-t')\}\langle[A(t),B(t')]_{\eta}\rangle, \quad (2)$$

where

$$[A,B]_{\eta}=AB-\eta BA,$$

$\eta=\pm 1$ (whichever is more convenient),

$$A(t)=e^{iHt}A(0)e^{-iHt},$$

$\theta(t)$ is the step function, unity for positive t and zero for negative t, and $\langle\cdots\rangle$ denote an average with respect to the canonical density matrix of the system at temperature T. We assume $\hbar=1$. In practice, it is convenient to work with the Fourier transform of the Green function with respect to ω:

$$\langle\langle A;B\rangle\rangle_{\omega}^{(\pm)}=\frac{1}{2\pi}\int_{-\infty}^{\infty}\langle\langle A(t);B(t')\rangle\rangle^{(\pm)}\times e^{-i\omega(t-t')}d(t-t'). \quad (3)$$

The Fourier transform satisfies the equation of motion[10]

$$\langle\langle A;B\rangle\rangle_{\omega}=(1/2\pi)\langle[A,B]_{\eta}\rangle+\langle\langle[A,H];B\rangle\rangle_{\omega}, \quad (4)$$

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where

$$\langle\langle A;B\rangle\rangle_\omega=\langle\langle A;B\rangle\rangle_\omega^{(+)}, \quad \text{if} \quad \mathrm{Im}\omega>0$$
$$=\langle\langle A;B\rangle\rangle_\omega^{(-)}, \quad \text{if} \quad \mathrm{Im}\omega<0 \qquad (5)$$

and $\mathrm{Im}\omega$ stands for the imaginary part of ω. It can be shown that[10]

$$\langle B(t')A(t)\rangle=i\lim_{\epsilon\to 0+}\int_{-\infty}^{\infty}\frac{\langle\langle A;B\rangle\rangle_{\omega+i\epsilon}-\langle\langle A;B\rangle\rangle_{\omega-i\epsilon}}{e^{\beta(\omega-\mu)}-\eta}\times e^{-i\omega(t-t')}d\omega, \qquad (6)$$

where $\beta=1/K_BT$ and K_B is the Boltzmann constant.

In our analysis, we consider the Green function

$$G_{jk}^{\sigma}(\omega)=\langle\langle c_{j\sigma};c_{k\sigma}^{\dagger}\rangle\rangle_\omega, \quad \eta=-1. \qquad (7)$$

The knowledge of this Green function enables one to evaluate the density of states per atom of spin σ, which is needed to study the condition for ferromagnetism. Substituting (7) into (6), putting $j=k$, $t-t'=0$, and summing over j, one obtains for $\langle n_\sigma\rangle$ the mean number of the electrons per atom of spin:

$$\langle n_\sigma\rangle=\frac{1}{N}\sum_j\langle c_{j\sigma}^{\dagger}c_{j\sigma}\rangle$$
$$=\frac{i}{N}\lim_{\epsilon\to 0+}\sum_j\int\frac{G_{jj}^{\sigma}(\omega+i\epsilon)-G_{jj}^{\sigma}(\omega-i\epsilon)}{e^{\beta(\omega-\mu)}+1}d\omega. \qquad (8)$$

This shows that

$$\rho_\sigma(\omega)=\frac{i}{N}\lim_{\epsilon\to 0+}\sum_j[G_{jj}^{\sigma}(\omega+i\epsilon)-G_{jj}^{\sigma}(\omega-i\epsilon)] \qquad (9)$$

gives the density of (pseudoparticle) states per atom of spin σ.

For the Hamiltonian (1), one finds

$$[c_{i\sigma},H]=\sum_j T_{ij}c_{j\sigma}+In_{i-\sigma}c_{i\sigma}+\sum_{j\sigma'}Q_{ij}n_{j\sigma'}c_{i\sigma} \qquad (10)$$

and

$$[n_{i\sigma},H]=\sum_j T_{ij}(c_{i\sigma}^{\dagger}c_{j\sigma}-c_{j\sigma}^{\dagger}c_{i\sigma}). \qquad (11)$$

Therefore, the Eq. (4) for $G_{ij}^{\sigma}(\omega)$ comes out to be

$$\omega G_{ik}^{\sigma}(\omega)=\frac{\delta_{ik}}{2\pi}+\sum_j T_{ij}G_{jk}^{\sigma}(\omega)+I\langle\langle n_{i-\sigma}c_{i\sigma};\rangle\rangle_\omega+\sum_{j\sigma'}Q_{ij}\langle\langle n_{j\sigma'}c_{i\sigma};\rangle\rangle_\omega. \qquad (12)$$

Here, and hereafter, the operator $c_{k\sigma}^{\dagger}$ coming after ";" in the Green functions is suppressed for brevity.

The right-hand side of Eq. (12) contains higher-order Green functions $\langle\langle n_{i-\sigma}c_{i\sigma};\rangle\rangle_\omega$ and $\langle\langle n_{j\sigma'}c_{i\sigma};\rangle\rangle_\omega$. To solve Eq. (12) for Green function $G_{ik}^{\sigma}(\omega)$, these higher-order Green functions are needed. For evaluating these higher-order Green functions, the equations of motion of the form (4) are written for these Green functions. These equations in turn involve still higher-order Green functions. To obtain the exact solution one has to solve a hierarchy of equations of the form (4). To get a plausible approximate solution one tries to truncate this hierarchy of equations at a certain stage by decoupling the higher-order Green functions into the lower-order Green functions. Accuracy of a solution depends on how many equations of motion of Green functions one solves and what scheme is resorted to for decoupling the higher-order Green functions.

We first examine the simple scheme, the Hartree-Fock approximation in which the higher-order Green functions appearing in (12) are decoupled as follows:

$$\langle\langle n_{i-\sigma}c_{i\sigma};\rangle\rangle_\omega\simeq\langle n_{i-\sigma}\rangle\langle\langle c_{i\sigma};\rangle\rangle_\omega,$$
$$\langle\langle n_{j\sigma'}c_{i\sigma};\rangle\rangle_\omega\simeq\langle n_{j\sigma'}\rangle\langle\langle c_{i\sigma};\rangle\rangle_\omega. \qquad (13)$$

In this approximation, Eq. (12) turns out to be

$$(\omega-I\langle n_{-\sigma}\rangle-QnZ)G_{ik}^{\sigma}(\omega)=\frac{\delta_{ik}}{2\pi}+\sum_j T_{ij}G_{jk}^{\sigma}(\omega). \qquad (14)$$

Here, Z is the number of nearest neighbors, and we have used the translational symmetry of the problem to write $\langle n_{i-\sigma}\rangle=\langle n_{-\sigma}\rangle$. We do not consider antiferromagnetic ordering. This equation may be solved by Fourier transformation. If we write

$$G_{ij}^{\sigma}(\omega)=\frac{1}{N}\sum e^{i\mathbf{k}\cdot(\mathbf{R}_i-\mathbf{R}_j)}G^{\sigma}(\mathbf{k},\omega) \qquad (15)$$

and use (A6), we obtain from (14)

$$G^{\sigma}(\mathbf{k},\omega)=(2\pi)^{-1}/(\omega-\epsilon_{\mathbf{k}}-I\langle n_{-\sigma}\rangle-QnZ). \qquad (16)$$

The poles of one-particle Green function give the energies of quasiparticles; Eq. (16), therefore, shows that the band structure is slightly modified, the energy of the $(\mathbf{k},\sigma)$ state now being $\epsilon_{\mathbf{k}}+I\langle n_{-\sigma}\rangle+QnZ$, which reduces to Hubbard's expression in the limit $Q\to 0$.

Now, we shall go beyond the Hartree-Fock approximation by writing down the equations of motion [Eq. (4)] for the higher-order Green functions $\langle\langle n_{i-\sigma}c_{i\sigma};\rangle\rangle_\omega$ and $\langle\langle n_{j\sigma'}c_{i\sigma};\rangle\rangle_\omega$. The equation of motion for $\langle\langle n_{i-\sigma}c_{i\sigma};\rangle\rangle_\omega$ is given by

$$(\omega-T_0-I)\langle\langle n_{i-\sigma}c_{i\sigma};\rangle\rangle_\omega$$
$$=\langle n_{-\sigma}\rangle\frac{\delta_{ik}}{2\pi}+\sum_{\substack{j\\ i\neq j}}T_{ij}\langle\langle n_{i-\sigma}c_{j\sigma};\rangle\rangle_\omega$$
$$+\sum_{\substack{j\\ i\neq j}}T_{ij}\{\langle\langle c_{i-\sigma}^{\dagger}c_{j-\sigma}c_{i\sigma};\rangle\rangle_\omega-\langle\langle c_{j-\sigma}^{\dagger}c_{i-\sigma}c_{i\sigma};\rangle\rangle_\omega\}$$
$$+\sum_{j\sigma'}Q_{ij}\langle\langle n_{i-\sigma}n_{j\sigma'}c_{i\sigma};\rangle\rangle_\omega, \qquad (17)$$

where

$$T_0=T_{ii}=\frac{1}{N}\sum_{\mathbf{k}}\epsilon_{\mathbf{k}}. \qquad (18)$$

We decouple the three-operator Green functions $\langle\langle n_{i-\sigma}c_{i\sigma};\rangle\rangle_\omega$, $\langle\langle c_{i-\sigma}^\dagger c_{j-\sigma}c_{i\sigma};\rangle\rangle_\omega$, and $\langle\langle c_{j-\sigma}^\dagger c_{i-\sigma}c_{i\sigma};\rangle\rangle_\omega$ according to the approximations used by Hubbard,[4] and five-operator Green function $\langle\langle n_{i-\sigma}n_{j\sigma'}c_{i\sigma};\rangle\rangle_\omega$ into $\langle\langle n_{i-\sigma}c_{i\sigma};\rangle\rangle_\omega$ according to the following approximation:

$$\langle\langle n_{i-\sigma}n_{j\sigma'}c_{i\sigma};\rangle\rangle_\omega \simeq \langle n_{\sigma'}\rangle\langle\langle n_{i-\sigma}c_{i\sigma};\rangle\rangle_\omega . \quad (19)$$

Within these approximations, Eq. (17) reduces to

$$\langle\langle n_{i-\sigma}c_{i\sigma};\rangle\rangle_\omega = \frac{\langle n_{-\sigma}\rangle}{\omega-T_0-I-QnZ}\left[\frac{\delta_{ik}}{2\pi}+\sum_{\substack{j\\ i\neq j}} T_{ij}G_{jk}^\sigma(\omega)\right]. \quad (20)$$

In a similar way, if we write the equation of motion for $\langle\langle n_{j\sigma'}c_{i\sigma};\rangle\rangle_\omega$ and decouple the three-operator Green functions according to the approximations used by Hubbard[4] and five-operator Green functions into $\langle\langle n_{j\sigma'}c_{i\sigma};\rangle\rangle_\omega$ according to the approximation used in Eq. (19), we get

$$\langle\langle n_{j\sigma'}c_{i\sigma};\rangle\rangle_\omega = \frac{\langle n_{\sigma'}\rangle}{\omega-T_0-I\langle n_{-\sigma}\rangle-QnZ}\left[\frac{\delta_{ik}}{2\pi}+\sum_{\substack{j\\ i\neq j}} T_{ij}G_{jk}^\sigma(\omega)\right]. \quad (21)$$

Substituting the values of Green functions $\langle\langle n_{i-\sigma}c_{i\sigma};\rangle\rangle_\omega$ and $\langle\langle n_{j\sigma'}c_{i\sigma};\rangle\rangle_\omega$ from Eqs. (20) and (21) in Eq. (12), we find

$$(\omega-T_0)G_{ik}^\sigma(\omega)=\{1+f(\omega,\langle n_{-\sigma}\rangle)\}\times\left\{\frac{\delta_{ik}}{2\pi}+\sum_{\substack{j\\ i\neq j}} T_{ij}G_{jk}^\sigma(\omega)\right\}, \quad (22)$$

where

$$f(\omega,\langle n_{-\sigma}\rangle)=\frac{I\langle n_{-\sigma}\rangle}{\omega-T_0-I-QnZ}+\frac{QnZ}{\omega-T_0-I\langle n_{-\sigma}\rangle-QnZ}. \quad (23)$$

The Fourier transform of the Green function defined by Eq. (15), is, therefore, given by

$$G^\sigma(\mathbf{k},\omega)=\frac{1}{2\pi}\frac{1+f(\omega,\langle n_{-\sigma}\rangle)}{(\omega-\epsilon_\mathbf{k})-(\epsilon_\mathbf{k}-T_0)f(\omega,\langle n_{-\sigma}\rangle)}. \quad (24)$$

We shall now consider the correlation effects in the theory of ferromagnetion on the basis of this Green function.

A. Approximate Solution in Zero-Bandwidth Limit

In the limit of zero bandwidth $\epsilon_\mathbf{k}=T_0$ for all $\mathbf{k}$, hence, it follows that $T_{ij}=T_0\delta_{ij}$. After replacing $\epsilon_\mathbf{k}$ by T_0 in Eq. (24) the Green function in zero-bandwidth case is

$$G^\sigma(\mathbf{k},\omega)=\frac{1}{2\pi}\frac{F(\omega)}{(\omega-T_0)(\omega-T_0-I-QnZ)(\omega-T_0-I\langle n_{-\sigma}\rangle-QnZ)}. \quad (25)$$

Here,

$$F(\omega)=\omega^2-(2T_0+I+QnZ)\omega+I^2\langle n_{-\sigma}\rangle(1-\langle n_{-\sigma}\rangle)-T_0(I+T_0+QnZ). \quad (26)$$

We substitute the value of $G^\sigma(\mathbf{k},\omega)$ from (25) in Eq. (9) to get the density of states per atom of spin σ,

$$\rho_\sigma(\omega)=\frac{I^2\langle n_{-\sigma}\rangle(1-\langle n_{-\sigma}\rangle)}{(I+QnZ)(I\langle n_{-\sigma}\rangle+QnZ)}\delta(\omega-T_0)+\frac{QnZ}{I\langle n_{-\sigma}\rangle+QnZ}\delta(\omega-T_0-I\langle n_{-\sigma}\rangle-QnZ)+\frac{I\langle n_{-\sigma}\rangle}{(I+QnZ)}\delta(\omega-T_0-I-QnZ). \quad (27)$$

This expression shows that system behaves as though it has three energy levels at T_0, $(T_0+I\langle n_{-\sigma}\rangle+QnZ)$, and $(T_0+I+QnZ)$ containing

$$I^2\langle n_{-\sigma}\rangle(1-\langle n_{-\sigma}\rangle)/(I+QnZ)(I\langle n_{-\sigma}\rangle+QnZ),$$

$QnZ/(I\langle n_{-\sigma}\rangle+QnZ)$, and $I\langle n_{-\sigma}\rangle/(I+QnZ)$ states per atom for spin σ, respectively. This result differs strikingly from that of Hubbard. According to Hubbard, the system behaves as though it has two energy levels at T_0 and T_0+I containing $(1-\langle n_{-\sigma}\rangle)$ and $\langle n_{-\sigma}\rangle$ states, respectively. It should be noted that because of interatomic interaction, the number of states in a particular energy level depends upon the strength of interaction parameters I and Q, while these are independent of the strength of interactions when interatomic interaction is neglected.

By finding out the density of states in each energy level, one can find out the number of electrons occupying a given energy level at absolute zero and, hence, the ground-state energy of the system. Comparing the energies of paramagnetic and ferromagnetic state, one can find out their relative stability. In the Hubbard model, it can be shown that for $n=1$, the energy of both paramagnetic and ferromagnetic states is the same and, hence, the probability of occurrence of both the states

is equal. In the present case, we find[11] that the energy of the ferromagnetic state is smaller than that of the paramagnetic state, and therefore the ferromagnetic state is more stable than the paramagnetic state.

B. Finite-Bandwidth Case

The general nature of the solution given by Eq. (24) will now be investigated for the finite-bandwidth case. The expression (24) for $G^\sigma(\mathbf{k},\omega)$ may be resolved into partial fractions:

$$G^\sigma(\mathbf{k},\omega)=\frac{1}{2\pi}\left[\frac{A_{\mathbf{k}\sigma}{}^{(1)}}{\omega-\omega_{\mathbf{k}\sigma}{}^{(1)}}+\frac{A_{\mathbf{k}\sigma}{}^{(2)}}{\omega-\omega_{\mathbf{k}\sigma}{}^{(2)}}+\frac{A_{\mathbf{k}\sigma}{}^{(3)}}{\omega-\omega_{\mathbf{k}\sigma}{}^{(3)}}\right]. \quad (28)$$

Here, $\omega_{\mathbf{k}\sigma}{}^{(1)}>\omega_{\mathbf{k}\sigma}{}^{(2)}>\omega_{\mathbf{k}\sigma}{}^{(3)}$ are the roots of the equation

$$\begin{aligned}(\omega-\epsilon_{\mathbf{k}})(\omega-T_0+I-QnZ)(\omega-T_0-I\langle n_{-\sigma}\rangle-QnZ)\\ -(\epsilon_{\mathbf{k}}-T_0)\{I\langle n_{-\sigma}\rangle(\omega-T_0-I\langle n_{-\sigma}\rangle-QnZ)\\ +2QnZ(\omega-T_0-I-QnZ)\}=0 \quad (29)\end{aligned}$$

and

$$A_{\mathbf{k}\sigma}{}^{(1)}=\frac{F(\omega_{\mathbf{k}\sigma}{}^{(1)})}{(\omega_{\mathbf{k}\sigma}{}^{(1)}-\omega_{\mathbf{k}\sigma}{}^{(2)})(\omega_{\mathbf{k}\sigma}{}^{(1)}-\omega_{\mathbf{k}\sigma}{}^{(3)})}, \quad (30)$$

$$A_{\mathbf{k}\sigma}{}^{(2)}=\frac{F(\omega_{\mathbf{k}\sigma}{}^{(2)})}{(\omega_{\mathbf{k}\sigma}{}^{(2)}-\omega_{\mathbf{k}\sigma}{}^{(1)})(\omega_{\mathbf{k}\sigma}{}^{(2)}-\omega_{\mathbf{k}\sigma}{}^{(3)})}, \quad (31)$$

$$A_{\mathbf{k}\sigma}{}^{(3)}=\frac{F(\omega_{\mathbf{k}\sigma}{}^{(3)})}{(\omega_{\mathbf{k}\sigma}{}^{(3)}-\omega_{\mathbf{k}\sigma}{}^{(1)})(\omega_{\mathbf{k}\sigma}{}^{(3)}-\omega_{\mathbf{k}\sigma}{}^{(2)})}. \quad (32)$$

From (28) and (9), the density of states per atom of spin σ is given by

$$\begin{aligned}\rho_\sigma(\omega)=\frac{1}{N}\sum_{\mathbf{k}}\{A_{\mathbf{k}\sigma}{}^{(1)}\delta(\omega-\omega_{\mathbf{k}\sigma}{}^{(1)})+A_{\mathbf{k}\sigma}{}^{(2)}\delta(\omega-\omega_{\mathbf{k}\sigma}{}^{(2)})\\ +A_{\mathbf{k}\sigma}{}^{(3)}\delta(\omega-\omega_{\mathbf{k}\sigma}{}^{(3)})\}. \quad (33)\end{aligned}$$

The expression (33) for density of states per atom of spin σ shows that the system behaves as though it has three bands with dispersion laws $\omega=\omega_{\mathbf{k}\sigma}{}^{(1)}$, $\omega=\omega_{\mathbf{k}\sigma}{}^{(2)}$, and $\omega=\omega_{\mathbf{k}\sigma}{}^{(3)}$. From Eqs. (30)–(32), it can be shown that $A_{\mathbf{k}\sigma}{}^{(1)}+A_{\mathbf{k}\sigma}{}^{(2)}+A_{\mathbf{k}\sigma}{}^{(3)}=1$. This equality and Eqs. (30)–(32) show that the effect of $A_{\mathbf{k}\sigma}{}^{(n)}$ cannot be given any simpler interpretation beyond the statement that they regulate the density of states in each band in such a way that the total number of states in all three bands is just one. The general form of the bands $\omega_{\mathbf{k}\sigma}{}^{(1)}$, $\omega_{\mathbf{k}\sigma}{}^{(2)}$, and $\omega_{\mathbf{k}\sigma}{}^{(3)}$ is shown in Fig. 1. The variation of bandwidth of different bands with Q is shown in Table I.

[11] Here and in what follows, for the purpose of calculation, we take $T_0=0$, i.e., we measure the band energy from the middle of the band, and consider a fcc lattice. For intra-atomic interaction parameter I, we take its screened (see Ref. 4) value which is equal to 10 eV. The screened value of interatomic interaction parameter is equal to 3 eV (see Ref. 4). But to study the dependence of interatomic correlation effects on the magnitude of interaction parameter Q, we vary Q from 0 to 3 eV.

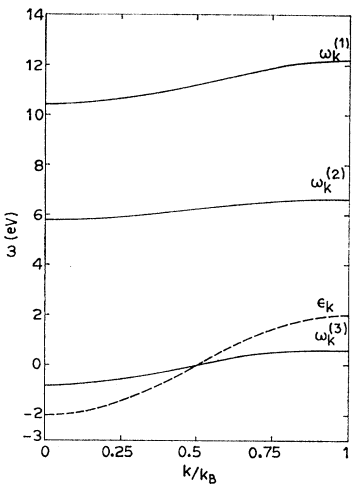

FIG. 1. Typical perturbed band structure is shown. $\omega=\omega_{k\sigma}{}^{(1)}$, $\omega=\omega_{k\sigma}{}^{(2)}$, and $\omega=\omega_{k\sigma}{}^{(3)}$ are three perturbed energy bands. Dashed curve represents the unperturbed band. k_B is the wave vector at the boundary of the Brillouin zone. The parameters corresponding to this figure are $I=10$ eV, $Q=0.1$ eV, $Z=12$, $\langle n_{-\sigma}\rangle=\frac{1}{2}n=0.5$.

We note that the bandwidth of lowest and highest bands increases as Q decreases, while bandwidth of middle band diminishes as Q decreases. This middle band disappears when $Q\to 0$.

Now, an expression more explicit than (33) will be given for the density of states. If $F(\omega_{\mathbf{k}\sigma}{}^{(1)})$ and $F(\omega_{\mathbf{k}\sigma}{}^{(3)})$ are positive and $F(\omega_{\mathbf{k}\sigma}{}^{(2)})$ is negative,[12] then by applying the property of δ function[13]

$$\delta(g(x))=\sum_n\frac{\delta(x-a_n)}{|g'(a_n)|}, \quad (34)$$

where a_n are the roots of the equation $g(x)=0$ and $|g'(a_n)|$ is the modulus of derivative of $g(x)$ with

TABLE I. Comparison of bandwidths of different bands for different values of Q.[a,b]

Q (eV)	Δ_1 (eV)	Δ_2 (eV)	Δ_3 (eV)
0.0	2.0	0.0	2.0
0.1	1.79	0.76	1.45
0.3	1.49	1.64	0.87
0.5	1.28	2.14	0.58
0.7	1.13	2.46	0.41
0.9	1.01	2.68	0.31
1.0	0.96	2.77	0.27
2.0	0.64	3.26	0.10
3.0	0.50	3.45	0.05

[a] Δ_1, Δ_2, and Δ_3 are the bandwidths of the bands $\omega=\omega_{\mathbf{k}\sigma}{}^{(1)}$, $\omega=\omega_{\mathbf{k}\sigma}{}^{(2)}$, and $\omega=\omega_{\mathbf{k}\sigma}{}^{(3)}$, respectively.
[b] Calculations are performed for $T_0=0$, $I=10$ eV, $Z=12$, $\langle n_{-\sigma}\rangle=\frac{1}{2}n=0.5$, and the bandwidth of the unperturbed band ($\omega=\epsilon_{\mathbf{k}}$) is taken equal to 4.0 eV.

[12] We have seen that this condition is actually satisfied.
[13] L. D. Landau and E. M. Lifshitz, *Quantum Mechanics* (Pergamon Press Inc., Oxford, 1965), p. 18.

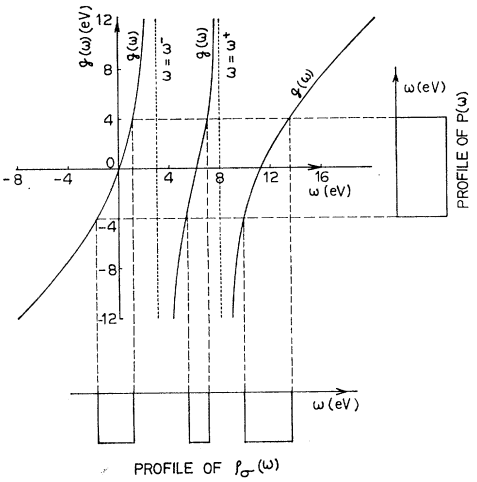

FIG. 2. Typical $g(\omega,\langle n_{-\sigma}\rangle)$ curve is shown. The projection of the unperturbed density-of-states function $P(\omega)$ into the perturbed density of states $\rho_\sigma(\omega)$ is indicated. The parameters corresponding to this figure are the same as in Fig. 1.

respect to x at a_n, the density of states can be written as

$$\rho_\sigma(\omega)=P\{g(\omega,\langle n_{-\sigma}\rangle)\}. \tag{35}$$

Here,

$$g(\omega,\langle n_{-\sigma}\rangle)=\frac{\omega+T_0+f(\omega,\langle n_{-\sigma}\rangle)}{1+f(\omega,\langle n_{-\sigma}\rangle)} \tag{36}$$

and

$$P(\omega)=\frac{1}{N}\sum_{\mathbf{k}}\delta(\omega-\epsilon_{\mathbf{k}}) \tag{37}$$

is the density of states corresponding to the band structure $\epsilon_{\mathbf{k}}$.

Thus, $\rho_\sigma(\omega)$ can be obtained from $P(\omega)$ by the transformation given by Eqs. (35) and (36). This transformation is illustrated graphically in Fig. 2 which shows a typical $g(\omega)$ curve and the projection of $P(\omega)$ into $\rho_\sigma(\omega)$. The curve $g(\omega,\langle n_{-\sigma}\rangle)$ splits into three parts. These parts are separated by infinities of $g(\omega,\langle n_{-\sigma}\rangle)$ at $\omega=\omega^+$, and $\omega=\omega^-$. The ω^+ and $\omega^-(\omega^+>\omega^-)$ are the roots of the equation

$$(\omega-T_0)(\omega-T_0-I-QnZ)+I^2\langle n_{-\sigma}\rangle(1-\langle n_{-\sigma}\rangle)=0. \tag{38}$$

TABLE II. Positions of infinities of $g(\omega,\langle n_{-\sigma}\rangle)$.[a]

Q (eV)	ω^+ (eV)	ω^- (eV)
0.0	5.0	5.0
0.1	8.12	3.08
0.5	14.24	1.76
1.0	20.80	1.20
2.0	33.25	0.75
3.0	45.45	0.55

[a] Calculations are performed for $T_0=0$, $I=10$ eV, $\langle n_{-\sigma}\rangle=\frac{1}{2}n=0.5$, and $Z=12$.

The values of ω^+ and ω^- are given in Table II for different values of Q. This Table illustrates that these infinities come closer as Q decreases and the width of the middle band is reduced. In the limit $Q\to 0$, both infinities merge together and the second band disappears. It is quite clear that the appearance of middle band is due to the effect of interatomic interaction within the decoupling scheme used here.

In the paramagnetic state ($\langle n_{-\sigma}\rangle=\langle n_\sigma\rangle=\frac{1}{2}n$) of the system, it is found by Hubbard that for square density of states, the first band is completely full for $n=1$ and, therefore, the system behaves as an insulator. We find that in the present case the system in the paramagnetic state behaves as an insulator only at some critical value $n=n_c$. The value of n_c depends on the value of Q. Table III shows the variation of n_c with Q. It is clear from the Table that n_c increases as Q decreases and it becomes equal to 1 as $Q\to 0$.

III. CONDITION FOR FERROMAGNETISM

In Sec. II, we have seen that in case of zero bandwidth, the ferromagnetic state is more stable than the paramagnetic state. In this section, we shall find out the possibility of the occurrence of ferromagnetism in the case of finite bandwidth. We use the general condition[4]

TABLE III. Critical value n_c for different values of Q.[a]

n_c (eV)	Q (eV)
1.0	0.0
0.84	0.1
0.52	0.5
0.35	1.0
0.21	2.0
0.16	3.0

[a] Calculation are performed for $T_0=0$, $I=10$ eV, $\langle n_{-\sigma}\rangle=\langle n_\sigma\rangle=\frac{1}{2}n$, $Z=12$, and the bandwidth of the unperturbed band equal to 4 eV.

$$-\tfrac{1}{2}\geq\int_{-\infty}^{\mu}\frac{\partial}{\partial n}[P\{g(\omega,\tfrac{1}{2}n)\}]d\omega, \tag{39}$$

which must be satisfied for a system to exist in the ferromagnetic state. Here, μ is the Fermi energy of the system.

It is difficult to manifest the condition (39) without reference to some specific density-of-states function $P(\omega)$. We examine the condition (39) for a square density of states

$$P(\omega)=1/\Delta, \quad \text{if } T^0-\tfrac{1}{2}\Delta<\omega<T^0+\tfrac{1}{2}\Delta$$
$$=0, \quad \text{otherwise.} \tag{40}$$

For this case, density of states $\rho_\sigma(\omega)$ can be written as

$$\rho_\sigma(\omega)=\frac{1}{\Delta}\sum_{\alpha=1}^{6}(-1)^{\alpha+1}\theta(\omega-\omega_\alpha{}^\sigma), \tag{41}$$

where $\omega_1^\sigma<\omega_3^\sigma<\omega_5^\sigma$ and $\omega_2^\sigma<\omega_4^\sigma<\omega_6^\sigma$ are the roots of the cubic equations

$$g(\omega,\tfrac{1}{2}n)=T_0-\tfrac{1}{2}\Delta \tag{42}$$

and

$$g(\omega,\tfrac{1}{2}n)=T_0+\tfrac{1}{2}\Delta, \tag{43}$$

respectively. The density of states given by Eq. (41), when used in Eq. (39), leads to the following condition for ferromagnetism:

$$-\tfrac{1}{2}\geqslant\frac{1}{\Delta}\int_{-\infty}^{\mu}\sum_{\alpha=1}^{6}(-1)^\alpha\frac{\partial\omega_\alpha{}^\sigma}{\partial n}\delta(\omega-\omega_\alpha{}^\sigma)d\omega. \tag{44}$$

It is not easy to evaluate right-hand side of inequality (44) analytically. We have evaluated it numerically.[14] It is found that when the Fermi level lies in the lowest band, ferromagnetism is not possible, but when it lies in the middle band, ferromagnetism is possible. This result is quite different from Hubbard's conclusion that for a square density of states ferromagnetism is not possible. One may, therefore, infer that the interatomic interaction should play an important role in determining the behavior of ferromagnetic metals.

IV. CONCLUSIONS

In the preceding sections, we have investigated an approximate model for electron correlation in an s band of transition metals. It is found that one band splits into three bands. The middle band occurs only because we consider the interatomic interactions. The system behaves as an insulator at certain critical value n_c of n. The value of n_c depends on Q. Conditions for the ferromagnetism for the square density of states are quite different from Hubbard's conclusion which is based on the assumption that interatomic interaction is negligible. We should mention here that Hubbard's decoupling approximation, which we have used, is not suitable for the Hamiltonian (1) which emphasizes intersite correlations. Also our expressions do not seem to preserve the electron-hole symmetry. However, our conclusions show that intersite correlations may be important to explain the magnetic properties of transition metals, and should not be neglected.

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APPENDIX

In this Appendix, we derive the approximate Hamiltonian of Eq. (1) for s band of the electrons. For an analysis of the ferromagnetism in transition metals we should have really considered the case of d electrons, but we choose to deal with s electrons only because of mathematical simplicity. However, while evaluating the various approximations which have gone into the derivation of the model, we shall keep in mind that we are dealing with $3d$ transition metals. One may expect that some important aspect of the real (d electrons) case will be missed in a study of the s-band case, but the analysis should be helpful in understanding the behavior of itinerant d electrons in ferromagnetic metals.

The dynamics of the s-band electrons may approximately be described by the Hamiltonian[4]

$$\begin{aligned}H=&\sum_{\mathbf{k}\sigma}\epsilon_{\mathbf{k}}c_{\mathbf{k}\sigma}{}^\dagger c_{\mathbf{k}\sigma}\\&+\tfrac{1}{2}\sum_{\substack{\mathbf{k}_1\mathbf{k}_2\mathbf{k}_1'\mathbf{k}_2'\\\sigma\sigma'}}\langle\mathbf{k}_1\mathbf{k}_2|\frac{1}{r}|\mathbf{k}_1'\mathbf{k}_2'\rangle c_{\mathbf{k}_1\sigma}{}^\dagger c_{\mathbf{k}_2\sigma'}{}^\dagger c_{\mathbf{k}_2'\sigma'}c_{\mathbf{k}_1'\sigma}\\&-\sum_{\substack{\mathbf{k}\mathbf{k}'\\\sigma}}\left\{2\langle\mathbf{k}\mathbf{k}'|\frac{1}{r}|\mathbf{k}\mathbf{k}'\rangle-\langle\mathbf{k}\mathbf{k}'|\frac{1}{r}|\mathbf{k}'\mathbf{k}\rangle\right\}\nu_{\mathbf{k}'}c_{\mathbf{k}\sigma}{}^\dagger c_{\mathbf{k}\sigma},\end{aligned} \tag{A1}$$

where $c_{\mathbf{k}\sigma}{}^\dagger$, $c_{\mathbf{k}\sigma}$ are the creation and annihilation operators of the electrons of wave vector $\mathbf{k}$ and spin σ, $\nu_{\mathbf{k}}$, is the occupation number of the state with wave vector $\mathbf{k}$ of the band in the Hartree-Fock calculation,

$$\begin{aligned}\langle\mathbf{k}_1\mathbf{k}_2|\frac{1}{r}|\mathbf{k}_1'\mathbf{k}_2'\rangle&\\=e^2\int\frac{\psi_{\mathbf{k}_1}(\mathbf{x})\psi_{\mathbf{k}_2}(\mathbf{x}')\psi_{\mathbf{k}_1'}(\mathbf{x})\psi_{\mathbf{k}_2'}(\mathbf{x}')}{|\mathbf{x}-\mathbf{x}'|}&d\mathbf{x}\,d\mathbf{x}',\end{aligned} \tag{A2}$$

$\epsilon_{\mathbf{k}}$ and $\psi_{\mathbf{k}}$ are the energy and wave function of the electron of wave vector $\mathbf{k}$ with some appropriate Hartree-Fock potential. It is assumed that the Hartree-Fock potential is independent of the electron spin so one has the same energy and wave function for both spins.

Now, we shall transform this Hamiltonian in Wannier representation. We transform the Bloch wave function into the Wannier function according to the relation

$$\psi_k(\mathbf{x})=\frac{1}{\sqrt{N}}\sum_i e^{-i\mathbf{k}\cdot\mathbf{R}_i}\phi(\mathbf{x}-\mathbf{R}_i), \tag{A3}$$

where $\phi(\mathbf{x}-\mathbf{R}_i)$ is the Wannier function at atomic position $\mathbf{R}_i$, and N is the total number of atoms in the system. Operators $c_{\mathbf{k}\sigma}{}^\dagger$ and $c_{\mathbf{k}\sigma}$ are transformed into the operators $c_{i\sigma}{}^\dagger$ and $c_{i\sigma}$ for an electron of spin σ in the

[14] Calculations are performed for $Q=0.1$, 1, and 3 eV, bandwidth $=4$ and 2 eV, and $0<n<1$.

orbital state $\phi(\mathbf{x}-\mathbf{R}_i)$ by the relations

$$c_{\mathbf{k}\sigma}^{\dagger}=\frac{1}{\sqrt{N}}\sum_i e^{i\mathbf{k}\cdot\mathbf{R}_i}c_{i\sigma}^{\dagger};\quad c_{\mathbf{k}\sigma}=\frac{1}{\sqrt{N}}\sum_i e^{-i\mathbf{k}\cdot\mathbf{R}_i}c_{i\sigma}. \tag{A4}$$

In terms of wave function $\phi(\mathbf{x}-\mathbf{R}_i)$ and operators $c_{i\sigma}^{\dagger}$, $c_{i\sigma}$, the Hamiltonian (A1) can be rewritten as

$$H=\sum_{ij\sigma} T_{ij}c_{i\sigma}^{\dagger}c_{j\sigma}+\tfrac{1}{2}\sum_{ijkl\sigma\sigma'}\langle ij|\frac{1}{r}|kl\rangle c_{i\sigma}^{\dagger}c_{j\sigma'}^{\dagger}c_{l\sigma'}c_{k\sigma}$$
$$-\frac{1}{N}\sum_{ijklm\sigma}\left\{2\langle ij|\frac{1}{r}|kl\rangle-\langle ij|\frac{1}{r}|lk\rangle\right\}\times\nu_{lj}c_{m\sigma}^{\dagger}c_{m+k-i\sigma}, \tag{A5}$$

where

$$T_{ij}=\frac{1}{N}\sum_{\mathbf{k}}\epsilon_k e^{i\mathbf{k}\cdot(\mathbf{R}_i-\mathbf{R}_j)}, \tag{A6}$$

$$\langle ij|\frac{1}{r}|kl\rangle=e^2\int\frac{\phi^*(\mathbf{x}-\mathbf{R}_i)\phi^*(\mathbf{x}'-\mathbf{R}_j)\phi(\mathbf{x}-\mathbf{R}_k)\phi(\mathbf{x}'-\mathbf{R}_l)}{|\mathbf{x}-\mathbf{x}'|}\times d\mathbf{x}d\mathbf{x}', \tag{A7}$$

and

$$\nu_{lj}=\frac{1}{N}\sum_k \nu_{\mathbf{k}}e^{i\mathbf{k}\cdot(\mathbf{R}_l-\mathbf{R}_j)}. \tag{A8}$$

One can evaluate the terms of the form given by Eq. (A7) by using atomic wave function, the representative magnitude of various terms for d electrons in transition metals is given by[4,15]

$$\begin{aligned}&\langle ii|1/r|ii\rangle\simeq 20\text{ eV},\\ &\langle ij|1/r|ij\rangle\simeq 6\text{ eV},\\ &\langle ii|1/r|ij\rangle\simeq \tfrac{1}{2}\text{ eV},\\ &\langle ij|1/r|ik\rangle\simeq \tfrac{1}{10}\text{ eV},\\ &\langle ii|1/r|jj\rangle\simeq 1/40\text{ eV},\end{aligned} \tag{A9}$$

where i, j, and k are all nearest neighbors. Other interaction terms which appear in Eq. (A5) are still smaller. From (A9), it is clear that the term $\langle ii|1/r|ii\rangle$ representing the intra-atomic correlation is the largest in magnitude. Hubbard retained only this term and neglected other terms. We note that the term $\langle ij|1/r|ij\rangle$ is about 30% of the term $\langle ii|1/r|ii\rangle$, and when we consider Z nearest neighbors which contribute equally, it does not appear convincing to neglect this term without a proper analysis. The other terms which are quite small may be neglected in comparison to these two terms. If we do so, we get from (A5)

$$H=\sum_{ij\sigma} T_{ij}c_{i\sigma}^{\dagger}c_{j\sigma}+\frac{I}{2}\sum_{i\sigma} n_{i\sigma}n_{i-\sigma}$$
$$+\tfrac{1}{2}\sum_{ij\sigma\sigma'} Q_{ij}n_{i\sigma}n_{j\sigma'}-n^2N\left[\frac{I}{2}+(N-1)Q\right], \tag{A10}$$

where

$$I=\langle ii|1/r|ii\rangle, \tag{A11}$$

$$Q_{ij}=\langle ij|1/r|ij\rangle=Q,$$
(if i and j are nearest neighbors)
$$=0,\quad\text{(otherwise)} \tag{A12}$$

n = number of electrons per atom.

After dropping out the last term in (A10) which is a constant, we obtain the Hamiltonian (1).

[15] J. L. Beeby, in *Theory of Magnetism in Transition Metals*, edited by W. Marshall (Academic Press Inc., New York, 1967), p. 87.