Ferromagnetism of an Electron Gas*

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In order to clarify whether or not the so-called itinerant-electron model can predict the existence of the ferromagnetism in the narrow d-band or transition metals, an electron gas in a background of a uniform positive charge (single-band model) is considered as a simplest model. On the basis of the interpolation formula for the correlation energy developed by Pines *et al.*, the difference of the energy between the ferromagnetic state and the unpolarized (paramagnetic) state is examined. Contrary to the prevailing view, apparently due to Wigner and Pines, if the dynamical screening for spin-polarized states is correctly taken into consideration, the electron gas then is shown to become ferromagnetic even in the intermediate-density region. The critical value of $(m^*/m)r_s$ [where (m^*/m) is the ratio of the band effective mass to the free mass of electrons, and r_s is the inter-electron spacing in units of the Bohr radius] beyond which the system becomes ferromagnetic lies between 7 and 10. The paramagnetic susceptibility also shows a divergence approximately over the same density range.

MUCH attention has been paid recently to the nature of ferromagnetism of the transition metals.[1] As starting points in studying this problem, two different approaches are generally employed. One is the so-called localized electron model in which the electrons pertinent to magnetism are taken to be localized near lattice points; the other is the itinerant (collective) electron model in which the electrons are assumed to be moving through the lattice as almost free electrons and to form various bands. If we use the latter model, the crucial point lies in how accurately one can treat the many-electron correlation effect. The correlation effect has been discussed to a certain extent, but the results are far from what one would call well established. In this paper we would like to emphasize the crucial importance of the dynamical screening effect in discussing the occurrence of ferromagnetism. By dynamical screening we mean the time dependence (retardation) of the effective screened Coulomb interaction which arises from the dielectric property of an electron gas as a screening medium; hence this screening is equivalent to the frequency dependence of the dielectric constant in an electron gas.[2] It is this effect that gives rise to a long-range interaction in the system through plasma oscillations, which becomes more important for the intermediate- or low-density electron gas. Because of our limited objective, we may formulate the problem on the basis of the simplest model, i.e., the single-band or Sommerfeld (jellium) model in which ions are assumed to be smeared out as a uniform background. Extension to the case of two degenerate bands is rather straightforward.

In the Hartree-Fock (HF) scheme in which only first-order exchange energy is considered, the energy of the system per particle in the paramagnetic state, $\epsilon_0(0)$, is given (in Rydberg units) by

$$\epsilon_0(0)=(3/\pi^2)(1/5\rho^2-1/2\rho)\,, \qquad (1)$$

where $\rho=\alpha r_s/\pi$, $\alpha=(4/9\pi)^{1/3}=0.521$, and r_s is defined as $(3/4\pi n)^{1/3}$ divided by the Bohr radius, n being the number density of electrons. Now let us consider polarized states of the system. We introduce a parameter to describe the degree of polarization, μ $(0\leqslant\mu\leqslant 1)$, and put the number of electrons with spin up and down equal to $n_+=(n/2)(1+\mu)$ and $n_-=(n/2)(1-\mu)$. The Fermi momentum in the polarized state, measured in units of the Fermi momentum in the unpolarized state, becomes

$$p_+=(1+\mu)^{1/3}, \quad p_-=(1-\mu)^{1/3}. \qquad (2)$$

Then the HF energy for polarized states is

$$\epsilon_0(\mu)=\frac{3}{2\pi^2}\left(\frac{(1+\mu)^{5/3}+(1-\mu)^{5/3}}{5\rho^2}-\frac{(1+\mu)^{4/3}+(1-\mu)^{4/3}}{2\rho}\right). \qquad (3)$$

A comparison of Eqs. (1) and (3) indicates that the (completely polarized) ferromagnetic state has lower energy than the paramagnetic state, $\epsilon_0(1)<\epsilon_0(0)$, when[3]

$$\rho>\tfrac{2}{5}(2^{1/3}+1) \quad \text{or} \quad r_s>5.45. \qquad (4)$$

On the other hand, partially polarized states have always higher energy than either the completely polarized state or the paramagnetic state and may be ignored.

We may not take the Bloch condition (4) as valid for actual metals because of the complete neglect of the correlation effect (or screening effect) in HF scheme. In fact, if we take the screening effect into account by the Thomas-Fermi potential, then it has been shown[4] that the paramagnetic state always remains the lowest state

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[1] For a review, see N. F. Mott, Advan. Phys. **13**, 325 (1964). Related references concerning the correlation effect are also found in this article.

[2] J. Lindhard, Kgl. Danske Videnskab. Selskab, Mat. Fys. Medd. **28**, No. 8 (1954).

[3] F. Bloch, Z. Physik **57**, 549 (1929).

[4] F. Iwamoto and K. Sawada, Phys. Rev. **126**, 887 (1962).

and the system can never become ferromagnetic. A more refined calculation[5] based on Landau's Fermi-liquid theory[6] has been performed, but the conclusion remains unchanged. However, it is important to note that in all these theories only static screening has been considered and the effect of dynamical screening has been completely ignored.

Here we shall rigorously show that, when the dynamical screening effect is preserved, the electron gas definitely becomes ferromagnetic for a density above a certain value of r_s. Our theory is in principle based on the interpolation formula of the correlation energy for the intermediate-density region developed by Pines, Hubbard, and Nozieres and Pines.[7] Taking Hubbard's result, however, we may conclude that the higher order exchange effects give a contribution of less than 10%[8] of the main term. Therefore, within this error, we may take the correlation energy in the form given by the random-phase approximation[9] [RPA; Eq. (38) in Hubbard's paper[7]]. In the Gell-Mann and Brueckner (GB) form[10] the correlation energy is[11]

$$\epsilon_c(\mu)=-\frac{3}{4\pi^5}\int_0^\infty\frac{dq}{q}\int_{-\infty}^\infty du \times\sum_{n=2}^\infty\frac{(-1)^n}{n}\left[\frac{Q_q^+(u)+Q_q^-(u)}{2}\right]^n\left(\frac{\rho}{\pi q^2}\right)^{n-2}, \quad (5)$$

where

$$Q_q^\pm(u)=\int_{\substack{|\mathbf{p}+\mathbf{q}|>p_\pm\\ p<p_\pm}} d^3p\int_{-\infty}^\infty dt \times\exp\left\{ituq-|t|\left(\frac{q^2}{2}+\mathbf{p}\cdot\mathbf{q}\right)\right\}. \quad (6)$$

The u dependence of $Q_q^\pm(u)$ reflects the dynamical screening effect. First of all, we consider the completely polarized ferromagnetic state for which $Q_q^-(u)$ in Eq. (5) vanishes. For this state, by applying a linear transformation

$$\mathbf{p}\to 2^{1/3}\mathbf{p},\quad \mathbf{q}\to 2^{1/3}\mathbf{q},$$
$$t\to 2^{-2/3}t,\quad u\to 2^{1/3}u,$$

we obtain

$$\epsilon_c(1)=-\frac{3}{4\pi^5}\left(\frac{1}{2}\right)\int_0^\infty\frac{dq}{q}\int_{-\infty}^\infty du \times\sum_{n=2}^\infty\frac{(-1)^n}{n}[Q_q(u)]^n\left(\frac{\rho}{2^{4/3}\pi q^2}\right)^{n-2}, \quad (7)$$

where $Q_q(u)$ is the function defined by Eq. (6) for the paramagnetic state ($p_\pm=1$). From this form it is seen that the correlation energy of the ferromagnetic state ($\mu=1$) is obtained from that of the paramagnetic state by replacing ρ by $\rho/2^{4/3}$ and dividing by 2; i.e.,

$$\epsilon_c(1;\rho)=\tfrac{1}{2}\epsilon_c(0;\rho/2^{4/3}). \quad (8)$$

Now let us take a sufficiently large value of ρ (low-density region) so that the analytic continuation $x^{-2}[x-\ln(1+x)]$ of the series

$$\sum_{n=2}^\infty\frac{(-1)^n}{n}x^{n-2}$$

which appears in Eq. (5) behaves like $1/x$ for large x.[12] This indicates that, for sufficiently large ρ, the correlation energy can always be approximated by the form

$$\epsilon_c(0)\approx -A/\rho, \quad (9)$$

where A is a positive numerical constant given by[13]

$$A=\frac{3}{4\pi^4}\int_0^\infty dq\, q\int_{-\infty}^\infty du\, Q_q(u)=\frac{3}{2\pi^2}. \quad (10)$$

It is important to note that the correlation energy of the form (9) is a natural consequence of the dynamical screening effect. When neglecting this effect, the value of A in Eq. (10) becomes indefinite and hence the energy can never take the form (9). In the light of Eq. (8), it is seen that the correlation energy of the paramagnetic state is definitely higher than that of the ferromagnetic state in this density region. Combining this with the

[5] S. Misawa, Phys. Letters 7, 249 (1963).

[6] L. D. Landau, Zh. Eksperim. i Teor. Fiz. 30, 1058 (1956) [English transl.: Soviet Phys.—JETP 3, 920 (1957)].

[7] D. Pines, in *Solid State Physics*, edited by F. Seitz and D. Turnbull (Academic Press Inc., New York, 1955), Vol. 1, p. 373; J. Hubbard, Proc. Roy. Soc. (London) A243, 336 (1957); P. Nozières and D. Pines, Phys. Rev. 111, 442 (1958).

[8] The Hubbard result for exchange correction to the correlation energy can be described by the formula $0.024-0.002r_s$ in the intermediate-density region. Since the first term independent of the density is unaltered by spin polarization, the energy difference between the para- and ferromagnetic state arises only from the second term; $0.002(1-2^{-1/3})r_s\approx 0.0004r_s$. This energy difference is less than 20% of the other term coming from the nonexchange correlation energy or the HF energy in the density region of interest ($r_s\sim 7$). It is also confirmed that the increment of this energy difference with respect to r_s is less than 10% of the increment arising from the other terms.

[9] Besides Ref. 7, D. Bohm and D. Pines, Phys. Rev. 92, 609 (1953); M. Gell-Mann and K. A. Brueckner, *ibid.* 106, 364 (1957); K. Sawada, K. A. Brueckner, N. Fukuda, and R. Brout, *ibid.* 106, 507 (1957).

[10] Ferrell has argued that the GB energy violates his convexity theorem for the energy when $r_s\gtrsim 1$ [R. A. Ferrell, Phys. Rev. Letters 1, 443 (1958)]. His validity criterion, however, has been obtained by taking only the first two terms of r_s expansion (high-density expansion). If, instead, we do keep the exact form of the GB energy (5), then their result never violates the convexity theorem for the whole range of r_s. This is in accordance with the fact that the mathematical structure of the GB energy in the low-density limit ($r_s\to\infty$) is exactly the same as that of Wigner's electron lattice [G. Iwata, Progr. Theoret. Phys. (Kyoto) 24, 1118 (1960)].

[11] K. A. Brueckner and K. Sawada, Phys. Rev. 112, 328 (1958).

[12] This procedure has also been justified by Iwata using a Mellin transform (see Ref. 10).

[13] Although the integral A apparently includes a divergent term of the form $\int d^3q\, v(q)$ [where $v(q)$ is the Coulomb potential], it can be shown that this term is completely cancelled with another divergence coming from the integration $\int_0^\infty dq\, q^3\int_{-\infty}^\infty du \times\ln[1+(\rho/\pi q^2)Q]$ for ϵ_c. Only the finite part of A determines the asymptotic behaviors of ϵ_c for large ρ, (9).

HF energy, which also gives lower energy for the ferromagnetic state when $\rho > \frac{2}{5}(2^{1/3}+1)$, we can conclude that above a certain critical value of ρ (or r_s), the ferromagnetic state has a lower energy than the paramagnetic state.[14]

The critical density where the transition from para- to ferromagnetic takes place is determined by a root of the equation

$$\epsilon_0(0)+\epsilon_c(0)-\epsilon_0(1)-\epsilon_c(1)=0. \tag{11}$$

To proceed in calculation we take the same approximate form for $Q_q(u)$ as used by GB[15];

$$\begin{aligned} Q_q(u) &= 4\pi[1-u\tan^{-1}(1/u)] \equiv 4\pi R(u) \quad \text{for} \quad 0 \leqslant q \leqslant 1, \\ &= 0 \quad \text{for} \quad q>1. \end{aligned} \tag{12}$$

This is justified by the fact that $Q_q(u)$ is appreciable only for small values of q and damps rapidly for large q, and also, in evaluating the correlation energy (5), contribution from integration over small q is predominant. In this approximation, the correlation energy in the paramagnetic state becomes

$$\epsilon_c(0) = \frac{3}{8\pi^2}\frac{1}{\rho^2}\int_0^\infty du \times \left\{ \ln(1+4\rho R) - (4\rho R)^2 \ln\left(1+\frac{1}{4\rho R}\right) - 4\rho R \right\}, \tag{13}$$

and for the ferromagnetic state [see Eq. (8)]

$$\epsilon_c(1) = \frac{3}{8\pi^2}\frac{2^{5/3}}{\rho^2}\int_0^\infty du \left\{ \ln(1+2^{2/3}\rho R) - (2^{2/3}\rho R)^2 \ln\left(1+\frac{1}{2^{2/3}\rho R}\right) - 2^{2/3}\rho R \right\}. \tag{14}$$

These energies have been evaluated numerically for various values of ρ and plotted in Fig. 1. From this result we obtain the final conclusion that the system becomes ferromagnetic when

$$r_s > 7.41. \tag{15}$$

We have assumed so far that the electron mass, in the absence of the Coulomb interaction, is equal to the bare electron mass *in vacuo*. In the case that the single-particle energy can be described in terms of a band effective mass m^* near the Fermi surface, the criterion (15) should be modified to read

$$(m^*/m)r_s > 7.41. \tag{16}$$

So far we have considered only the completely polarized state. However, if partially polarized states are taken into account, then the critical condition (15) or (16) might be modified so that the partially polarized ferromagnetic state is first established for a lower value of r_s. By observing the singular point of the paramagnetic susceptibility χ, it will be shown that this is not the case. From the μ^2 term in the energy, we obtain, in the ratio to the noninteracting value χ_0,

$$\chi/\chi_0 = [1-\rho+s(\rho)]^{-1}, \tag{17}$$

where the second term comes from HF and the third term, in the approximation (12),

$$s(\rho) = \frac{4\rho^2}{\pi}\int_0^\infty du \frac{R(u)}{(1+u^2)^2} \ln\left(1+\frac{1}{4\rho R(u)}\right) \tag{18}$$

represents the contribution from correlation. Since $s(\rho)>0$, the correlation effect always reduces the susceptibility in this approximation,[16] but is not so great

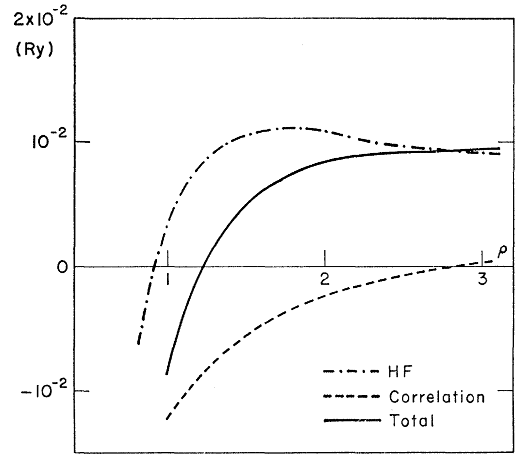

FIG. 1. Energy difference between paramagnetic and ferromagnetic states plotted against $\rho(=\alpha r_s/\pi \approx r_s/6)$. For $\rho>1.23$ ($r_s>7.41$) the ferromagnetic state becomes energetically favorable.

[14] This conclusion seems to contradict Pines (see Ref. 7) who claims that there is no possibility of ferromagnetism. However, he has used the assumption that the change in screening due to spin polarization may be neglected. This is not correct, but instead the change in screening is crucial, in our view, for the occurrence of ferromagnetism. In particular, the relation (8) cannot be obtained by Pine's assumption.

[15] Since the δ term (constant in r_s) in the GB energy is ignored, this approximation is not good for the high-density region ($r_s \lesssim 2$). However, for the intermediate density region in which we are interested ($5 \lesssim r_s \lesssim 10$) the approximation seems to be very good; this is confirmed by comparing our results with Hubbard's values (see Ref. 7) which have been numerically evaluated without approximation.

[16] This statement should not be taken as general, since the approximation (12) is not so accurate for this case. In fact, taking the low-density limit of the correlation energy (5), we have

$$\epsilon_c(\mu) \sim -\frac{3}{4\pi^4}\frac{1}{\rho}\int_0^\infty dq\, q\int_{-\infty}^\infty du \frac{Q_q^+(u)+Q_q^-(u)}{2} = -\frac{(1+\mu)^{4/3}+(1-\mu)^{4/3}}{2}\frac{A}{\rho}$$

as the exact value, where A was given by Eq. (10). Therefore, in this limit, $s(\rho)$ should be

$$s(\rho) \sim -(2\pi^2/3)A\rho,$$

which is negative, while the approximate expression (18) always gives positive values.

as to reverse the enhancement due to the HF term. In fact, because of the inequality $\ln[1+1/x]<1/x$ for $x>0$,

$$s(\rho)<\frac{\rho}{\pi}\int_0^\infty \frac{du}{(1+u^2)^2}=\frac{\rho}{4}, \tag{19}$$

and

$$\chi_0/\chi<1-\tfrac{3}{4}\rho. \tag{20}$$

Therefore, again, the paramagnetic state is locally unstable with respect to the spin polarization for ρ greater than $\frac{4}{3}$. A more accurate value for this critical point is found by numerical integration of $s(\rho)$ to be $\rho=1.25$ ($r_s=7.51$), which is very close to but slightly larger than the critical point of (15) or (16).[17]

{*Note added in proof.* The results obtained above are somewhat sensitive to the accuracy with which we have treated the effect of higher order exchange terms (exchange correlation). In preceding discussions we have examined this on the basis of Hubbard's calculation.[8] However, his scheme[7] is not consistent with the physical requirement that the pair distribution function for particles of like spin should vanish at the origin. To satisfy this requirement, it is essential to take the second-order exchange term with the dynamically screened Coulomb interaction [see S. Ueda, Progr. Theoret. Phys. (Kyoto) **26**, 45 (1961)]. This exchange-correlation energy, denoted by $\epsilon_c^{ex}(\mu;r_s)$, can be written down as three 4-dimensional energy-momentum integrations of an integrand consisting of the product of four one-particle propagators, one bare Coulomb potential, and one dynamically screened Coulomb potential. Using this fact we can derive the relation of ϵ_c^{ex} between the ferromagnetic state and the paramagnetic state:

$$\epsilon_c^{ex}(1;r_s)=\epsilon_c^{ex}(0;r_s/2^{4/3}). \tag{21}$$

The explicit evaluation of ϵ_c^{ex} as a function of r_s is very difficult, so we will estimate this from the results of Nozières' and Pines' analysis.[7] The energy ϵ_c^{ex} consists of two parts, one arising from the short-range part of the screened interaction, the other coming from the long-range part; i.e.,

$$\epsilon_c^{ex}(0;r_s)=\frac{0.046}{1+0.326\beta^2}+0.0136\frac{\beta^4}{r_s}, \tag{22}$$

where β is the usual cutoff parameter of the plasma oscillations. As a function of r_s, β for the paramagnetic state may be determined by Ferrell's relation [R. A. Ferrell, Phys. Rev. **107**, 450 (1957)]

$$\frac{\alpha r_s}{\pi}=\frac{\beta^2}{(2+\beta)\ln(1+2/\beta)-2}. \tag{23}$$

From Eqs. (21), (22), and (23), we can calculate the energy difference, $\epsilon_c^{ex}(1;r_s)-\epsilon_c^{ex}(0;r_s)$, between the ferromagnetic state and the paramagnetic state. The results are 0.0047, 0.0049, and 0.0052 Ry for $r_s=8$, 10, and 12, respectively. If we make these corrections to Eq. (11), then the critical value of r_s in (15) is shifted to approximately nine. If we further correct the error introduced by the approximation (12), it is safe to conclude that, within the scope of the present treatment, the system becomes ferromagnetic when

$$(m^*/m)r_s \gtrsim 10. \tag{24}$$ }

Finally, we will make a comment concerning a requirement which more refined theories must satisfy. From the structure of Eq. (13) or (14) it is seen that the most important term in the correlation energy is always of the form $-A/r_s$. This r_s^{-1} term corresponds to the leading term in the usual low-density expansion. It is remarkable that even in the intermediate density region, $1\lesssim r_s\lesssim 10$, the most important term is given in this form. We should note that in order to obtain this form it is essential to make the analytically continued function from a complete series of the perturbation expansion. This kind of situation must be preserved in more refined theories. It is never reasonable to describe physical quantities in terms of the "bare" Coulomb interaction. We should always take the dynamically screened Coulomb interaction as the real interaction of physical meaning; this is in accordance with the fact that this screened interaction itself is obtained as a result of analytic continuation.

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[17] This difference should not be taken very seriously, because of the approximation (see Ref. 16).