PHYSICAL REVIEW B VOLUME 7, NUMBER 1 1 JANUARY 1973

Spin Polarization of a Ferromagnetic Electron Gas Due to a Finite Concentration of Impurities[*]

D. J. Kim
Francis Bitter National Magnet Laboratory,[*] *Massachusetts Institute of Technology,*
Cambridge, Massachusetts 02139
and Physics Department, Aoyama Gakuin University,[†] *Chitosedal, Setagaya-Ku, Tokyo, Japan*
(Received 14 February 1972)

Spin polarization of an interacting electron gas due to a random distribution of magnetic impurities is discussed by going beyond the ordinary linear approximation. The effect of the nonlinearity is obtained in the form of modifying the wave-number-dependent magnetic susceptibility so as to include a change in the spin splitting of the electron energy bands proportional to the impurity concentration.

I. INTRODUCTION

Recently, motivated by the elastic diffuse neutron-scattering experiments[1] which give us detailed information on the spatial behavior of the host-metal spin polarization around an impurity, we discussed the charge and spin susceptibilities of an electron gas at the ferromagnetic state.[2,3] In actual analysis of the neutron data, for instance, Fe in Pd,[4] however, we encounter a kind of nonlinear problem. Namely, the observed conduction-electron spin polarization of Pd due to a finite concentration of Fe impurities is not reproduced simply by the product of the spin susceptibility of the pure paramagnetic Pd and the molecular magnetic field due to the impurities. Thus, we argued that the susceptibility to be used in calculating the conduction-electron spin polarization in Pd is not the susceptibility of the pure Pd but the susceptibility of the Pd spin split by the impurities.[5]

The purpose of the present paper is to reformulate our earlier discussion of Ref. 5 along the line of Ref. 3 (referred to as I). In the process of this reformulation we make some important improvements on the earlier work summarized in Sec. III.

II. INTERACTING ELECTRON GAS WITH MANY IMPURITIES

The Hamiltonian of an interacting electron gas is the same as I and it is the sum of the kinetic energy and the Coulomb repulsion:

$$\mathcal{H}^0=\sum_{l,\sigma}\epsilon_l c_{l\sigma}^\dagger c_{l\sigma}+\frac{1}{2}\sum_{\substack{l,l',\kappa\\ \sigma,\sigma'}}V(\kappa)c_{l\sigma}^\dagger c_{l'\sigma'}^\dagger c_{l'-\kappa,\sigma'}c_{l+\kappa,\sigma}\ , \tag{2.1}$$

where $c_{l\sigma}^\dagger$ is the creation operator of an electron with energy ϵ_l and spin $\sigma=\pm$, $V(\kappa)$ is the Fourier transform of the Coulomb interaction, $4\pi e^2/\kappa^2$, and we assume the unit volume for the system.

The electron gas described by the Hamiltonian equation (2.1) can be either ferromagnetic or paramagnetic depending upon the temperature and the values of the parameters appearing in the Hamiltonian. As in I, this part of the problem is handled by the Hartree–Fock or random-phase-approximation (RPA) procedure.[6] In either case, ferromagnetic or paramagnetic, the conduction-electron spin density is uniform in the unperturbed state. Our problem is to see how the conduction-electron spin polarization is disturbed if we introduce a finite number, say, N_0, of magnetic impurities into the electron gas.

The actual interaction between the conduction electrons and impurities can be complicated but here we consider the impurities only as sources of molecular magnetic field. Namely, in using the s-d exchange interaction model we retain only the longitudinal or z component. This seems to be justified, since in this paper we assume always that the impurity spins are ferromagnetically ordered. Under these assumptions the effective magnetic field $H(r)$, or its Fourier transform $H(\kappa)$, is given as

$$\mu_B H(\kappa)=(J(\kappa)/N)\langle S^z\rangle\sum_i e^{i\kappa R_i}\ , \tag{2.2}$$

where $\langle S^z\rangle$ is the magnitude of a ferromagnetically ordered impurity spin, $J(\kappa)$ is the s-d exchange integral, N is the total number of lattice points in the system, and R_i is the site of the impurity. The Zeeman energy of the conduction electrons, which is nothing but the longitudinal component of the s-d exchange interaction, is given in terms of $H(\kappa)$ as

$$\mathcal{H}_m'=-\mu_B\sum_{l,\kappa}H(\kappa)(c_{l+}^\dagger c_{l-\kappa,+}-c_{l-}^\dagger c_{l-\kappa,-})\ . \tag{2.3}$$

Our problem is to calculate the conduction-electron number density for each spin,

$$n_\pm(q)=\left\langle\sum_k c_{k\pm}^\dagger c_{k+q,\pm}\right\rangle\ , \tag{2.4}$$

under the Hamiltonian $\mathcal{H}^0+\mathcal{H}_m'$ using the double-time Green's function.[7] The necessary Green's functions are of the form of $\langle c_{k+q,\pm}|c_{k,\pm}^\dagger\rangle_\omega$, from which the electron number density defined in Eq. (2.4) is obtained by the following well-known procedure:

$$n_\pm(q)=\sum_k-\frac{1}{\pi}\,\mathrm{Im}\int_{-\infty}^{\infty}\langle c_{k+q,\pm}|c_{k\pm}^\dagger\rangle_{\omega+i0^+}f(\omega)\,d\omega\ , \tag{2.5}$$

where $f(\omega)$ is the Fermi distribution function. The important thing to note is that the thermal average $\langle\cdots\rangle$ is to be taken for a certain given spatial distribution of impurities and that we do not average over the random distribution of impurities. We take this attitude since we want to use our result in discussing the cross section of magnetic diffuse scattering of neutrons in dilute ferromagnetic alloys where the average over the impurity distribution is to be taken in $[n_\pm(q)]^2$ and not in $n_\pm(q)$.[8]

The equation of motion for the Green's function under the Hamiltonian $\mathcal{H}^0+\mathcal{H}_m'$ is easily obtained, as in I:

$$(\omega-\epsilon_{k+q})\langle c_{k+q,\pm}|c_{k\pm}^\dagger\rangle_\omega=\delta_{q,0}\mp\sum_\kappa \mu_B H(\kappa)\langle c_{k+q-\kappa,\pm}|c_{k,\pm}^\dagger\rangle_\omega+\sum_{l,\kappa,\sigma'}V(\kappa)\langle c_{l,\sigma'}^\dagger c_{l-\kappa,\sigma'}c_{k+q+\kappa,\pm}|c_{k\pm}^\dagger\rangle_\omega\,, \tag{2.6}$$

where we take the chemical potential as the origin of measuring energy. The first approximation is to introduce the RPA to the last Coulomb interaction terms of Eq. (2.6). By the procedure shown in Eqs. (2.20)–(2.23) of I, Eq. (2.6) is reduced to

$$\begin{aligned}[\omega-\epsilon_{k+q}-\tilde{V}(0)n_\pm(0)]\langle c_{k+q,\pm}|c_{k,\pm}^\dagger\rangle_\omega=\delta_{q,0}\mp\sum_\kappa \mu_B H(\kappa)\langle c_{k+q-\kappa,\pm}|c_{k,\pm}^\dagger\rangle_\omega+V(q)[n_+(q)+n_-(q)]\langle c_{k\pm}|c_{k\pm}^\dagger\rangle_\omega(1-\delta_{q,0})\\ -\tilde{V}(q)n_\pm(q)\langle c_{k\pm}|c_{k\pm}^\dagger\rangle_\omega(1-\delta_{q,0})\,,\end{aligned} \tag{2.7}$$

where $\tilde{V}(q)$ is the effective exchange interaction defined as

$$\sum_\kappa V(\kappa)\langle c_{k-\kappa,\pm}^\dagger c_{k+q-\kappa,\pm}\rangle\equiv\tilde{V}(q)\sum_k\langle c_{k-\kappa,\pm}^\dagger c_{k+q-\kappa,\pm}\rangle\,. \tag{2.8}$$

Note that, as discussed in I, $\tilde{V}(q)$ does not diverge for $q\to 0$, unlike the Coulomb interaction $V(q)$.

The equations of the form of Eq. (2.7) for different q's form a closed set of equations which, in principle, can be solved exactly. The actual solution of Eq. (2.7), however, is not simple. Thus, as the second approximation, we retain only the term proportional to $H(q)$ in the second term on the right-hand side of Eq. (2.7). It is clear other terms with $\kappa\neq q$ are of the higher orders. In this approximation Eq. (2.7) is reduced to

$$\begin{aligned}(\omega-\epsilon_{k+q,\pm})\langle c_{k+q,\pm}|c_{k\pm}^\dagger\rangle_\omega&=\delta_{q,0}\mp\mu_B H(q)\langle c_{k\pm}|c_{k\pm}^\dagger\rangle_\omega\\ &\quad+V(q)[n_+(q)+n_-(q)]\langle c_{k\pm}|c_{k\pm}^\dagger\rangle_\omega(1-\delta_{q,0})\\ &\quad-\tilde{V}(q)n_\pm(q)\langle c_{k\pm}^\dagger|c_{k\pm}\rangle_\omega(1-\delta_{q,0})\,,\end{aligned} \tag{2.9}$$

where we introduced

$$\epsilon_{k,\pm}\equiv\epsilon_k-\tilde{V}(0)n_\pm(0)\,. \tag{2.10}$$

Note $\epsilon_{k+q,\pm}$ depends on the impurity concentration through the term $n_\pm(0)$, as will be seen below.

We solve Eq. (2.9) exactly, not by the linear approximation of I, as

$$\langle c_{k\pm}|c_{k\pm}^\dagger\rangle_\omega=\frac{1}{\omega-\epsilon_{k\pm}\pm\mu_B H(0)+i0^+}\,, \tag{2.11}$$

$$\begin{aligned}\langle c_{k+q,\pm}|c_{k\pm}^\dagger\rangle_\omega&=\mp\mu_B H(q)\frac{1}{\omega-\epsilon_{k\pm}+i0^+}\,\frac{1}{\omega-\epsilon_{k+q,\pm}+i0^+}\\ &\quad+V(q)[n_+(q)+n_-(q)]\frac{1}{\omega-\epsilon_{k\pm}+i0^+}\,\frac{1}{\omega-\epsilon_{k+q,\pm}+i0^+}\\ &\quad-\tilde{V}(q)n_\pm(q)\frac{1}{\omega-\epsilon_{k\pm}+i0^+}\,\frac{1}{\omega-\epsilon_{k+q,\pm}+i0^+}\,.\end{aligned} \tag{2.12}$$

By inserting Eqs. (2.11) and (2.12) into Eq. (2.5) the electron density for each spin is obtained as

$$n_\pm(0)=\sum_\kappa f(\epsilon_{k\pm}\mp\mu_B H(0))\,, \tag{2.13}$$

$$n_\pm(q)=\pm\frac{\tilde{F}_\pm(q)+2V(q)\tilde{F}_+(q)\tilde{F}_-(q)}{1+V(q)[\tilde{F}_+(q)+\tilde{F}_-(q)]}\,\mu_B H(q)\,, \tag{2.14}$$

where

$$\tilde{F}_\pm(q)=F_\pm(q)/[1-\tilde{V}(q)F_\pm(q)]\,, \tag{2.15}$$

$$F_\pm(q)=-\sum_k\frac{f(\epsilon_{k\pm})-f(\epsilon_{k+q,\pm})}{\epsilon_{k\pm}-\epsilon_{k+q,\pm}}\,. \tag{2.16}$$

The result of Eq. (2.14) can be rewritten by introducing a susceptibility $\chi^{\mathrm{mm}}(q)$ as

$$\begin{aligned}\mu_B[n_+(q)-n_-(q)]&\equiv M(q)\\ &=\chi^{\mathrm{mm}}(q)H(q)\\ &=\chi^{\mathrm{mm}}(q)\left(\frac{1}{\mu_B}\,\frac{1}{N}\,J(q)\langle S^z\rangle\sum_i e^{iqR_i}\right)\,,\end{aligned} \tag{2.17}$$

where

$$\chi^{\mathrm{mm}}(q)=\mu_B^2\,\frac{\tilde{F}_+(q)+\tilde{F}_-(q)+4V(q)\tilde{F}_+(q)\tilde{F}_-(q)}{1+V(q)[\tilde{F}_+(q)+\tilde{F}_-(q)]}\,. \tag{2.18}$$

Formally the result contained in Eqs. (2.14)–(2.18) is very similar to the corresponding result of I, but the present result is different from I in a very essential point. The difference comes from the difference between $\epsilon_{k\pm}$ of I and the present paper [Eq. (2.26) of I and Eq. (2.10) of the present paper]. This difference causes the differences in $f(\epsilon_{k\pm})$, $F_\pm(q)$, $F_\pm(q)$, and $\chi^{\mathrm{mm}}(q)$ of I and the present paper. For instance, $\chi^{\mathrm{mm}}(q)$ appearing in Eq. (2.17) is not the susceptibility of the pure electron gas as obtained in I but the susceptibility of the electron gas which includes the effect of the impurities, as we discuss below.

Thus, the prescription to calculate the conduction-electron spin polarization is summarized as follows: (i) From a self-consistent treatment of Eq. (2.13) we determine $n_\pm(0)$ as a function of the impurity concentration. (ii) We calculate $\chi^{mm}(q)$ for the bands whose spin splitting is given by the $n_\pm(0)$ through Eq. (2.10), and this is the susceptibility to be used in Eq. (2.17).

Let us see how $\chi^{mm}(q)$ of Eq. (2.17) depends on the impurity concentration. According to prescription (ii) the susceptibility should be calculated for bands spin split by an amount of $\tilde{V}(0)[n_-(0) - n_+(0)]$ and this splitting can be rewritten as

$$\tilde{V}(0)[n_-(0) - n_+(0)] \cong \tilde{V}(0)(n_-^0 - n_+^0) + \tilde{V}(0)\left(\chi_0^{mm}(0)\frac{-H(0)}{\mu_B}\right)$$
$$= \tilde{V}(0)(n_-^0 - n_+^0) - \frac{N_0}{N}\frac{1}{\mu_B^2} J(0)\langle S^z\rangle \tilde{V}(0)\chi_0^{mm}(0), \qquad (2.19)$$

where $\chi_0^{mm}(0)$ is the uniform susceptibility of the pure electron gas with spin splitting $\tilde{V}(0)(n_-^0 - n_+^0)$ [Eq. (3.3) of I] and $n_\pm^0$ are the number of electrons with ± spins in the unperturbed state. Note that in the strongly magnetic electron gas, such as Pd metals, $\tilde{V}(0)\chi_0^{mm}(0)$ is close to 1. Generally $\tilde{V}(0)\chi_0^{mm}(0)$ is not much less than 1. Anyway, Eq. (2.19) shows that in calculating $\chi^{mm}(q)$ to be used in Eq. (2.17) an additional spin splitting proportional to the impurity concentration must be taken into account. This is the procedure we used in analyzing the neutron data on PdFe alloys.[5]

III. DISCUSSION

The earlier result of Ref. 5 and the present result are essentially the same in emphasizing the necessity of considering the additional spin splitting of the bands due to the presence of impurities. But there are two differences between these works. One is the difference in the models. In this paper we took into account the long-range nature of the Coulomb interaction for the reason discussed in I. In Ref. 5 we used the Hubbard-type model, which is now known to be inappropriate in calculating $\chi^{mm}(q)$.

Another difference is the way of approximating the second term on the right-hand side of Eq. (2.7). In this paper we retained only one term proportional to $H(q)$ for all q including the case of $q = 0$, whereas in Ref. 5 we retained one more term which is proportional to $H(0)$ for the case of $q \neq 0$. Either one of these procedures is an approximation, and it is not easy to say which is better. However, with the earlier approximation of Ref. 5 we had the difficulty of $\lim_{q\to 0} M(q) \neq M(0)$. This difficulty is removed in the present paper.

ACKNOWLEDGMENTS

The author thanks Dr. B. B. Schwartz and Dr. H. C. Praddaude for valuable discussions.

---

*Supported by The National Science Foundation.

†Present address.

[1] For a review, see G. G. Low, Advan. Phys. 18, 371 (1969).

[2] D. J. Kim, H. C. Praddaude, and B. B. Schwartz, Phys. Rev. Letters 23, 419 (1969).

[3] D. J. Kim, B. B. Schwartz, and H. C. Praddaude, preceding paper, Phys. Rev. B 7, 205 (1972), hereafter referred to as I.

[4] G. G. Low and T. M. Holden, Proc. Phys. Soc. (London) 89, 119 (1966).

[5] D. J. Kim and B. B. Schwartz, Phys. Rev. Letters 21, 1744 (1968).

[6] T. Izuyama, D. J. Kim, and R. Kubo, J. Phys. Soc. Japan 18, 1025 (1963).

[7] D. N. Zubarev, Usp. Fiz. Nauk 71, 71 (1960) [Sov. Phys. Usp. 3, 320 (1960)].

[8] W. Marshall, J. Phys. C 1, 88 (1968).