

Zero-Temperature Susceptibility of a Localized Spin Exchange Coupled with the Conduction Electrons

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A zero-temperature magnetic susceptibility of a localized spin antiferromagnetically exchange-coupled with the conduction electrons is calculated on the basis of the model in which a singlet bound state is formed between the localized spin and the conduction electrons. The obtained susceptibility is given by $\mu_B^2/|\tilde{E}|$ where \tilde{E} denotes the binding energy. It is shown that this result holds in any stage of approximation.

§ 1. Introduction

For a system consisting of the conduction electrons and a localized spin which are coupled by the antiferromagnetic exchange interaction, $J < 0$, Yosida¹⁾ (henceforth, referred to as I) has shown by using a modified perturbation method that a singlet bound state appears in the ground state of this system. Okiji²⁾ confirmed this conclusion by performing the higher order calculations. Further, it has been shown³⁾ that the energy of this ground state with the singlet bound state is lower by the binding energy than that of the normal state and also some extensions of this theory have been done.

Since these calculations are done at the absolute zero of temperature and in the absence of a magnetic field, it would be a relevant problem as a next step to extend this theory to finite temperatures and non-zero magnetic fields. The effect of a static magnetic field acting only on the localized spin, for example, is considered qualitatively as follows. The singlet bound state is composed with the same weight of the two spin components of the localized spin whose magnitude is one-half. The magnetic field changes this ratio, so that the magnetic moment is induced to the ground state and spin flip which is essential to gain the binding energy will become difficult as the magnetic field is increased. A critical field at which the bound state disappears may be of the same order as the binding energy of $H=0$, but it is now difficult to treat this problem.

In this paper we focus ourselves on the limit of weak field and calculate the magnetic susceptibility of the bound state at 0°K. First, we consider, for simplicity, the case in which the magnetic field only interacts with the localized

spin. After that we shall show that the case where the conduction electrons also see the magnetic field can be treated without any essential change from the former case except the Pauli paramagnetism. In connection with the calculation of the susceptibility, we shall add the discussion of the triplet state in the case of the antiferromagnetic interaction, in which a non-realistic bound state appears at and after the first approximation.

§ 2. Calculation

We consider the effect of a magnetic field applied to the system consisting of the conduction electrons and a localized spin which are coupled by the antiferromagnetic exchange interaction. First we assume that only the localized spin interacts with the magnetic field. The Hamiltonian is given by

$$H = H_0 + V + H_z, \quad (1)$$

$$H_0 = \sum_{k\sigma} \epsilon_k a_{k\sigma}^* a_{k\sigma}, \quad (2)$$

$$V = -\frac{J}{2N} \sum_{kk'} [(a_{k'\uparrow}^* a_{k\uparrow} - a_{k'\downarrow}^* a_{k\downarrow}) S_z + a_{k'\uparrow}^* a_{k\downarrow} S_- + a_{k'\downarrow}^* a_{k\uparrow} S_+], \quad (3)$$

$$H_z = g\mu_B H S_z, \quad (4)$$

where H_z represents the Zeeman energy of the localized spin, g its g -factor, μ_B the Bohr magneton and H the magnetic field applied along the z -axis. Other notations are the same as in the previous papers.^{1), 2), 3)} The magnitude of the localized spin is assumed to be one-half.

The wave function of the ground state is expanded as follows:

$$\begin{aligned} \phi = & [\sum_k (F_k^\alpha a_{k\downarrow}^* \alpha + F_k^\beta a_{k\uparrow}^* \beta) \\ & + \sum_{k_1 k_2 k_3} (F_{k_1 k_2 k_3}^{\alpha\downarrow} a_{k_1\downarrow}^* a_{k_2\downarrow}^* a_{k_3\downarrow} \alpha + F_{k_1 k_2 k_3}^{\beta\uparrow} a_{k_1\uparrow}^* a_{k_2\uparrow}^* a_{k_3\uparrow} \beta \\ & + F_{k_1 k_2 k_3}^{\alpha\uparrow} a_{k_1\downarrow}^* a_{k_2\uparrow}^* a_{k_3\uparrow} \alpha + F_{k_1 k_2 k_3}^{\beta\downarrow} a_{k_1\uparrow}^* a_{k_2\downarrow}^* a_{k_3\downarrow} \beta) + \dots] \phi_v, \end{aligned} \quad (5)$$

where α and β , respectively, denote the spin-up and spin-down state of the localized spin and ϕ_v represents the state of the Fermi sea. Inserting the expressions (2), (3), (4) and (5) into the Schrödinger equation

$$(H - E)\phi = 0, \quad (6)$$

we set up the simultaneous equations for the coefficients, F , in the same way as in the case $H=0$.¹⁾ We notice here that the eigenvalues of H_z for the α - and β -components of the wave function ϕ are $g\mu_B H/2$ and $-g\mu_B H/2$, respectively. That is, the Schrödinger equation (6) can be expressed as

$$0 = (H_0 + V + \mathcal{A} - E)\phi_\alpha + (H_0 + V - \mathcal{A} - E)\phi_\beta, \quad (7)$$

where

$$\phi_\alpha = |\alpha\rangle\langle\alpha|\psi\rangle, \phi_\beta = |\beta\rangle\langle\beta|\psi\rangle, \text{ and } \Delta = g\mu_B H/2. \quad (8)$$

Therefore, in the presence of the magnetic field, we can derive an equation for Γ from that of $\Delta=0$ ¹⁾ by replacing $-E$ by $\Delta-E$ in the coefficients of $\Gamma^\alpha, \Gamma^{\alpha\downarrow}, \Gamma^{\alpha\uparrow}, \dots$ and by $-\Delta-E$ in those of $\Gamma^\beta, \Gamma^{\beta\uparrow}, \Gamma^{\beta\downarrow}, \dots$. The calculation can be performed in parallel to the case of $\Delta=0$ and the details are omitted. Eliminating Γ_{k,k_2,k_3} from the equations, we obtain as the first approximation the following equations which correspond to Eqs. (19) and (20) of I:

$$\begin{aligned} & \Gamma_k^\alpha \left[\varepsilon_k + \Delta - E - 2 \left(\frac{J}{4N} \right)^2 \sum_{\mu\nu} \left(\frac{1}{\varepsilon_\mu + \varepsilon_k - \varepsilon_\nu + \Delta - E} + \frac{2}{\varepsilon_\mu + \varepsilon_k - \varepsilon_\nu - \Delta - E} \right) \right] \\ & + \frac{J}{4N} \sum_\mu (\Gamma_\mu^\alpha - 2\Gamma_\mu^\beta) + \left(\frac{J}{4N} \right)^2 \sum_{\mu\nu} \left(\frac{\Gamma_\mu^\alpha + 2\Gamma_\mu^\beta}{\varepsilon_\mu + \varepsilon_k - \varepsilon_\nu + \Delta - E} + \frac{2\Gamma_\mu^\beta}{\varepsilon_\mu + \varepsilon_k - \varepsilon_\nu - \Delta - E} \right) = 0, \end{aligned} \quad (9)$$

$$\begin{aligned} & \Gamma_k^\beta \left[\varepsilon_k - \Delta - E - 2 \left(\frac{J}{4N} \right)^2 \sum_{\mu\nu} \left(\frac{2}{\varepsilon_\mu + \varepsilon_k - \varepsilon_\nu + \Delta - E} + \frac{1}{\varepsilon_\mu + \varepsilon_k - \varepsilon_\nu - \Delta - E} \right) \right] \\ & + \frac{J}{4N} \sum_\mu (\Gamma_\mu^\beta - 2\Gamma_\mu^\alpha) + \left(\frac{J}{4N} \right)^2 \sum_{\mu\nu} \left(\frac{2\Gamma_\mu^\alpha}{\varepsilon_\mu + \varepsilon_k - \varepsilon_\nu + \Delta - E} + \frac{\Gamma_\mu^\beta + 2\Gamma_\mu^\alpha}{\varepsilon_\mu + \varepsilon_k - \varepsilon_\nu - \Delta - E} \right) = 0. \end{aligned} \quad (10)$$

In these equations, the shifts of the kinetic energy ε_k can be calculated as

$$\begin{aligned} & - 2 \left(\frac{J}{4N} \right)^2 \sum_{\mu\nu} \left(\frac{1}{\varepsilon_\mu + \varepsilon_k - \varepsilon_\nu + \Delta - E} + \frac{2}{\varepsilon_\mu + \varepsilon_k - \varepsilon_\nu - \Delta - E} \right) \\ & \simeq - 2 \left(\frac{J\rho}{4N} \right)^2 \left[6D \log 2 + (-E + \Delta) \log \frac{-E + \Delta}{D} + 2(-E - \Delta) \log \frac{-E - \Delta}{D} \right], \end{aligned} \quad (11)$$

$$\begin{aligned} & - 2 \left(\frac{J}{4N} \right)^2 \sum_{\mu\nu} \left(\frac{2}{\varepsilon_\mu + \varepsilon_k - \varepsilon_\nu + \Delta - E} + \frac{1}{\varepsilon_\mu + \varepsilon_k - \varepsilon_\nu - \Delta - E} \right) \\ & \simeq - 2 \left(\frac{J\rho}{4N} \right)^2 \left[6D \log 2 + 2(-E + \Delta) \log \frac{-E + \Delta}{D} + (-E - \Delta) \log \frac{-E - \Delta}{D} \right], \end{aligned} \quad (12)$$

where a squared state density is assumed and ρ and D express its constant state density and the half of the band width. The common term, $-12D(J\rho/4N)^2 \log 2$, to Eqs. (11) and (12) is interpreted as the energy shift of the bottom of the scattering state, which is obtained by the usual perturbation, and can be renormalized³⁾ to the energy as

$$\tilde{E} = E + 12D \left(\frac{J\rho}{4N} \right)^2 \log 2. \quad (13)$$

We neglect other terms, $(J\rho/4N)^2 (-E \pm \Delta) \log [(-E \pm \Delta)/D]$, because they are smaller than $(-E \pm \Delta)$ by the order, $(J\rho/4N)$. Then, Eqs. (9) and (10) can be written as

$$\Gamma_k^\alpha = -\frac{J}{4N} \frac{1}{\varepsilon_k - \tilde{E} + A} [(G^\alpha - 2G^\beta) + f_\alpha(\varepsilon_k)], \quad (14)$$

$$\Gamma_k^\beta = -\frac{J}{4N} \frac{1}{\varepsilon_k - \tilde{E} - A} [(G^\beta - 2G^\alpha) + f_\beta(\varepsilon)], \quad (15)$$

where

$$G^\alpha = \sum_k \Gamma_k^\alpha, \quad G^\beta = \sum_k \Gamma_k^\beta, \quad (16)$$

$$f_\alpha(\varepsilon) = \frac{J}{4N} \sum_{\mu\nu} \left(\frac{\Gamma_\mu^\alpha + 2\Gamma_\mu^\beta}{\varepsilon_\mu + \varepsilon - \varepsilon_\nu - \tilde{E} + A} + \frac{2\Gamma_\mu^\beta}{\varepsilon_\mu + \varepsilon - \varepsilon_\nu - \tilde{E} - A} \right), \quad (17)$$

$$f_\beta(\varepsilon) = \frac{J}{4N} \sum_{\mu\nu} \left(\frac{2\Gamma_\mu^\alpha}{\varepsilon_\mu + \varepsilon - \varepsilon_\nu - \tilde{E} + A} + \frac{\Gamma_\mu^\beta + 2\Gamma_\mu^\alpha}{\varepsilon_\mu + \varepsilon - \varepsilon_\nu - \tilde{E} - A} \right). \quad (18)$$

By using the method of successive approximation the simultaneous integral equations (14) and (15) can be reduced to the equations for G^α and G^β , up to the third order in J as

$$\begin{aligned} 0 = G^\alpha &+ \frac{J\rho}{4N} \int_0^D \frac{d\varepsilon}{\varepsilon - \tilde{E} + A} (G^\alpha - 2G^\beta) - \left(\frac{J\rho}{4N} \right)^3 \int_0^D \frac{d\varepsilon}{\varepsilon - \tilde{E} + A} \int_0^D d\varepsilon_1 \int_{-D}^0 d\varepsilon_2 \\ &\times \left[\frac{1}{\varepsilon + \varepsilon_1 - \varepsilon_2 - \tilde{E} + A} \left[\frac{G^\alpha - 2G^\beta}{\varepsilon_1 - \tilde{E} + A} + \frac{2(G^\beta - 2G^\alpha)}{\varepsilon_1 - \tilde{E} - A} \right] + \frac{2(G^\beta - 2G^\alpha)}{(\varepsilon + \varepsilon_1 - \varepsilon_2 - \tilde{E} - A)(\varepsilon_1 - \tilde{E} - A)} \right], \end{aligned} \quad (19)$$

$$\begin{aligned} 0 = G^\beta &+ \frac{J\rho}{4N} \int_0^D \frac{d\varepsilon}{\varepsilon - \tilde{E} - A} (G^\beta - 2G^\alpha) - \left(\frac{J\rho}{4N} \right)^3 \int_0^D \frac{d\varepsilon}{\varepsilon - \tilde{E} - A} \int_0^D d\varepsilon_1 \int_{-D}^0 d\varepsilon_2 \\ &\times \left[\frac{1}{\varepsilon + \varepsilon_1 - \varepsilon_2 - \tilde{E} - A} \left[\frac{G^\beta - 2G^\alpha}{\varepsilon_1 - \tilde{E} - A} + \frac{2(G^\alpha - 2G^\beta)}{\varepsilon_1 - \tilde{E} + A} \right] + \frac{2(G^\alpha - 2G^\beta)}{(\varepsilon + \varepsilon_1 - \varepsilon_2 - \tilde{E} + A)(\varepsilon_1 - \tilde{E} + A)} \right]. \end{aligned} \quad (20)$$

The integrals in Eqs. (19) and (20) are calculated under the condition of $|\tilde{E}| > 3A$ by retaining the logarithmic term of the highest order and the above two equations can be written as

$$0 = G^\alpha (1 - x + x^3 - 2x^2y - 2xy^2 + \frac{2}{3}y^3) + G^\beta (2x - x^3 + x^2y + xy^2 - \frac{1}{3}y^3), \quad (21)$$

$$0 = G^\alpha (2y - \frac{1}{3}x^3 + x^2y + xy^2 - y^3) + G^\beta (1 - y + \frac{2}{3}x^3 - 2x^2y - 2xy^2 + y^3), \quad (22)$$

where

$$x = \frac{J\rho}{4N} \log \frac{-\tilde{E} + A}{D}, \quad y = \frac{J\rho}{4N} \log \frac{-\tilde{E} - A}{D}. \quad (23)$$

From the condition that the simultaneous Eqs. (21) and (22) have a non-trivial solution, we obtain the secular equation

$$0 = F(x, y) \equiv (2 + x^3 - 3x^2y - 3xy^2 + y^3)^2 - (x^3 - 3x - 1)(y^3 - 3y - 1). \quad (24)$$

If we put $\Delta = 0$, x becomes equal to y and Eq. (24) is factorized as

$$F(x) \equiv (1 - 3x - 3x^3)(1 + x - \frac{5}{3}x^3) = 0. \quad (25)$$

The solution of the equation

$$0 = 1 - 3x - 3x^3 \quad (26)$$

gives the singlet bound state of $x = 0.305$ and that of the equation

$$0 = 1 + x - \frac{5}{3}x^3 \quad (27)$$

gives the triplet bound state of $x = 1.08$.¹⁾ This triplet bound state did not appear at the zeroth approximation¹⁾ and it is, in this stage of approximation, a false solution, because the present perturbation expansion is considered to be not convergent for a triplet state. We shall discuss this point in some details in the Appendix. There is a possibility that the magnetic field Δ will mix this 'wrong' triplet bound state in the singlet bound state, so that Eq. (24) gives no correct answer for the large value of Δ . However, for the infinitesimal Δ , we may use Eq. (24) to see how the singlet bound state varies with the magnetic field. We expand \tilde{E} , x and y , respectively, about the values of $\Delta = 0$ as follows:

$$\tilde{E} = \tilde{E}_0 + \delta\tilde{E}, \quad (28)$$

$$x = x_0 + \delta x, \quad \delta x = \frac{J\rho}{4N} \left[\frac{\delta\tilde{E} - \Delta}{\tilde{E}_0} - \frac{(\delta\tilde{E} - \Delta)^2}{2\tilde{E}_0^2} + \dots \right], \quad (29)$$

$$y = x_0 + \delta y, \quad \delta y = \frac{J\rho}{4N} \left[\frac{\delta\tilde{E} + \Delta}{\tilde{E}_0} - \frac{(\delta\tilde{E} + \Delta)^2}{2\tilde{E}_0^2} + \dots \right], \quad (30)$$

where x_0 is the solution of Eq. (26) and \tilde{E}_0 is related to x_0 as $x_0 = (J\rho/4N) \log(-\tilde{E}_0/D)$. Substituting Eqs. (29) and (30) into Eq. (24), we obtain, neglecting the higher order terms with respect to $J\rho/4N$,

$$F(x_0) + \frac{1}{2} \frac{d}{dx_0} F(x_0) \cdot (\delta x + \delta y) = 0. \quad (31)$$

Putting $F(x_0) = 0$ and $F'(x_0) \neq 0$, we obtain

$$\tilde{E} = \tilde{E}_0 + \frac{\Delta^2}{2\tilde{E}_0} \left[1 + O\left(\frac{J\rho}{4N}\right) \right]. \quad (32)$$

In the zeroth approximation which is given by neglecting the third power of x and y in Eq. (24), the same result as Eq. (31) is obtained, where the value of \tilde{E}_0 is taken to be that of the zeroth approximation, $\tilde{E}_0 = -D \exp(4N/3J\rho)$.

The functional form of Eq. (31) indicates that the energy of the singlet

bound state in a weak magnetic field is generally given by Eq. (32), namely, in each stage of the approximation, in which \tilde{E}_0 is the value at its approximation. Thus, the result of Eq. (32) is exact in so far as the exact value is used for \tilde{E}_0 .

This result can be proved more directly. Also in the higher order perturbation, simultaneous equations for G^α and G^β can be expressed in terms of x and y , in so far as the logarithmic terms of the highest order are retained. Instead of calculating the exact form of the secular equation $F(x, y) = 0$, we can use its Taylor expansion about $x=y=x_0$ for a small value of Δ ,

$$y = x_0 + y'(x_0)(x - x_0) + \frac{1}{2} y''(x_0)(x - x_0)^2 + \dots \quad (33)$$

$y'(x_0)$ is estimated as follows. $F(x, y)$ is symmetric with respect to x and y , because of the invariance under the inversion of the magnetic field direction. Furthermore, $F(x, y)$ is given by a polynomial of x and y , so that it is expressed as

$$0 = F(x, y) = \sum_{mn} A_{mn} (x^m y^n + x^n y^m).$$

Differentiating this with respect to x and setting $x=y=x_0$, we obtain $y'(x_0) = -1$ if $F(x, y)$ has no double root at $x=y=x_0$ or

$$\sum_{mn} (m+n) A_{mn} x_0^{m+n-1} = F'(x_0) \neq 0.$$

Then Eq. (33) becomes

$$y = 2x_0 - x + \frac{1}{2} y''(x_0)(x - x_0)^2 + \dots \quad (34)$$

Substituting Eqs. (29) and (30), we obtain

$$\tilde{E} = \tilde{E}_0 + \frac{\Delta^2}{2\tilde{E}_0} \left[1 + \frac{J\rho}{8N} y''(x_0) \right] + \dots \quad (35)$$

This is just the same expression as Eq. (32). From this we obtain the magnetic susceptibility χ ,

$$\chi = -\frac{\partial^2 \tilde{E}}{\partial H^2} = -\frac{1}{\tilde{E}_0} \left(\frac{1}{2} g \mu_B \right)^2. \quad (36)$$

To justify this conclusion we must show that the 'wrong' triplet bound state stated before has no influence on Eq. (36). The contribution of the triplet bound state to the result of Eq. (35) is estimated by the second order perturbation of H_z ,

$$E_{st} = \frac{|\langle \phi^t | H_z | \phi^s \rangle|^2}{\tilde{E}_0 - \tilde{E}_t}, \quad (37)$$

where ϕ^s and ϕ^t denote the wave function of singlet and triplet bound states,

respectively, and \tilde{E}_t is the energy of the triplet bound state. Using the wave function and the energy of the first approximation, we estimate E_{st} as

$$E_{st} = a \left(\frac{\tilde{E}_t}{\tilde{E}_0} \log^2 \frac{\tilde{E}_t}{\tilde{E}_0} \right) \frac{\Delta^2}{\tilde{E}_0}, \quad (38)$$

where a is the constant of the order unity. E_{st} is smaller than $\Delta^2/2\tilde{E}_0$ in Eq. (34) by a factor $(\tilde{E}_t/\tilde{E}_0) \log^2(\tilde{E}_t/\tilde{E}_0)$. In the limit $J \rightarrow 0$ this factor tends to zero, so that the triplet bound state has little effect on the result and the contribution to $\Delta^2/2\tilde{E}_0$ seems to come from triplet scattering states.

For triplet scattering states we use the following wave function as a crude approximation,

$$\begin{aligned} \phi_k^t &= \frac{1}{\sqrt{2}} (a_{k\downarrow}^* \alpha + a_{k\uparrow}^* \beta) \psi_0, \\ \phi_{k_1 k_2 k_3}^{t_1} &= \frac{1}{\sqrt{2}} (a_{k_1\downarrow}^* a_{k_2\downarrow}^* a_{k_3\downarrow} \alpha + a_{k_1\uparrow}^* a_{k_2\uparrow}^* a_{k_3\uparrow} \beta) \psi_0, \\ \phi_{k_1 k_2 k_3}^{t_2} &= \frac{1}{\sqrt{2}} (a_{k_1\downarrow}^* a_{k_2\uparrow}^* a_{k_3\uparrow} \alpha + a_{k_1\uparrow}^* a_{k_2\downarrow}^* a_{k_3\downarrow} \beta) \psi_0, \end{aligned} \quad (39)$$

and calculate the perturbed energy of the singlet bound state up to the order J^2 as

$$\begin{aligned} \sum_k \frac{|\langle \phi_k^t | H_z | \psi^s \rangle|^2}{\tilde{E}_0 - \epsilon_k} &= \frac{\Delta^2}{2\tilde{E}_0}, \\ \sum_{k_1 k_2 k_3} \frac{|\langle \phi_{k_1 k_2 k_3}^{t_1} | H_z | \psi^s \rangle|^2 + |\langle \phi_{k_1 k_2 k_3}^{t_2} | H_z | \psi^s \rangle|^2}{\tilde{E}_0 - (\epsilon_{k_1} + \epsilon_{k_2} - \epsilon_{k_3})} &= \frac{\Delta^2}{2\tilde{E}_0} \left(\frac{J\rho}{4N} \right)^2 \frac{15}{2(1+3x_0^2)}. \end{aligned} \quad (40)$$

The first expression is the same as that of Eq. (32). Thus, we can see that the magnetic susceptibility of the singlet bound state results almost from the transition to the triplet scattering states with one electron excitation. As we have seen, Eq. (32) holds in general. It is to be noted here that the change of the distribution of the conduction electrons gives rise to only an effect of $1/N$ compared with (36), as can be seen from Eq. (42).

Next we consider the case where the conduction electrons also interact with the magnetic field. For simplicity it is assumed that the g -value of the conduction electron is the same as that of the localized spin. H_z in Eq. (4) is replaced by

$$H_z = g\mu_B H [S_z + \frac{1}{2} \sum_k (a_{k\uparrow}^* a_{k\uparrow} - a_{k\downarrow}^* a_{k\downarrow})]. \quad (41)$$

The wave function is given by Eq. (5) with a modification of the region of k

in the summand, because of the polarization of the conduction band. The shift of the Fermi surface is denoted by η . In order to clarify a distinction between the wave functions without and with the polarization, we denote them as ϕ and ϕ' , respectively. Then, the kinetic energy of the electron in the up band is $\varepsilon_k - \eta$: $0 \leq \varepsilon_k \leq D + \eta$ or approximately as $0 \leq \varepsilon_k \leq D$, because η is negligible against D . Also that of the down band is $\varepsilon_k + \eta$: $0 \leq \varepsilon_k \leq D$, and the energies of hole states are considered in a similar way. Using the above, we can write the Schrödinger equation as

$$\begin{aligned} (H_0 + H_z + V - E)\phi' &\simeq (H_0 + \eta + \rho\eta^2 - 2\rho\eta\Delta + V + \frac{J\rho}{2N}\eta - E)\phi_\alpha \\ &+ (H_0 - \eta + \rho\eta^2 - 2\rho\eta\Delta + V - \frac{J\rho}{2N}\eta - E)\phi_\beta. \end{aligned} \quad (42)$$

Comparing Eq. (42) with Eq. (7), we notice that $\eta(1 + J\rho/2N)$ corresponds to Δ in Eq. (7). The constant term $-(\rho\eta^2 - 2\rho\eta\Delta)$ is the energy gain of the conduction electron system due to the magnetic field and Pauli paramagnetism is derived from it. Thus the energy shift η is equal to Δ . Therefore, there is no difference between Eq. (42) and Eq. (7) besides Pauli paramagnetism. In this case χ is given by

$$\chi = \chi_p - \frac{1}{\tilde{E}} \left(\frac{1}{2} g \mu_B \right)^2. \quad (43)$$

Here $\eta(J\rho/2N)$ is neglected with respect to η in Eq. (42).

§ 3. Discussion

We have obtained above a constant zero-temperature susceptibility, which is given by $\mu_B^2/|\tilde{E}_0| \sim \mu_B^2/kT_c$. $|\tilde{E}_0|$ is the binding energy which will tend to $-D \exp(N/J\rho)$. Thus, it may be expected that the magnetic susceptibility of the localized spin increases monotonically as temperature is lowered and approaches the above constant value, saturating at low temperatures if we assume a smooth change in temperature as has been asserted by Suhl and Wong.⁴⁾

A constant zero-temperature susceptibility has been obtained by Takano and Ogawa⁵⁾ and also by Dworin⁶⁾ who uses the Anderson model. However, Dworin's value is proportional to

$$\chi_D \sim \frac{J\rho}{N} \frac{\mu_B^2}{kT_c}$$

and is smaller by a factor of $J\rho/N$ than ours. Therefore, his value for the binding energy of the singlet bound state seems to be larger by the same factor than ours.

Recently, Hamann⁷⁾ has succeeded in solving Nagaoka's coupled integral equations and shown that the susceptibility diverges at the absolute zero of

temperature, although the magnitude of the localized spin vanishes for $S=1/2$. This result is certainly contradictory to our expectation. However, in order to elucidate this point, we must extend our theory to finite temperatures.

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Appendix

It has been mentioned in the text that for the antiferromagnetic exchange, a non-realistic triplet bound state appears at and after the first approximation in the perturbation expansion, and this makes it difficult to treat the effect of the magnetic field comparable to the binding energy \tilde{E} . In this Appendix, we shall briefly discuss a possibility for a triplet bound state on the basis of the simplest approximation. For ferromagnetic exchange, the same equation for $x = (J\rho/4N)\log(|\tilde{E}|/D)$ expressed by a power series with respect to x has no definite root within the radius of convergence.^{1), 2)} This seems to indicate that for a triplet state the present perturbation expansion does not lead us to a convergent result. Therefore, in order to avoid this difficulty in treating the effect of a magnetic field, we must solve the problem in a closed form.

The simplest approximation for obtaining the binding energy may be to restrict the excited states to those states with only one excited electron-hole pair. Even in such a case, it is still difficult to obtain an exact solution. Therefore, we shall here return to Eqs. (24), (25) and (26) of I and discuss a triplet bound state. These equations can be written for a triplet state as

$$\Gamma_k = \frac{J}{4N} \frac{1}{\varepsilon_k - \tilde{E}} [G + f(\varepsilon_k)], \quad (\text{A1})$$

$$G = \sum_k \Gamma_k, \quad (\text{A2})$$

$$f(\varepsilon_k) = -5 \frac{J}{4N} \sum_{\mu\nu} \frac{\Gamma_\mu}{\varepsilon_\mu + \varepsilon_k - \varepsilon_\nu - \tilde{E}}. \quad (\text{A3})$$

The iteration used in I gives the following series which determines the binding energy :

$$\begin{aligned} 1 = & \frac{\rho J}{4N} \int_0^D \frac{d\varepsilon}{\varepsilon - \tilde{E}} \left[1 + 5 \left(\frac{J\rho}{4N} \right)^2 \int_0^D \frac{\mathcal{Y}(\varepsilon_1 + \varepsilon)}{\varepsilon_1 - \tilde{E}} d\varepsilon_1 \right. \\ & \left. + 5^2 \left(\frac{J\rho}{4N} \right)^4 \int_0^D \frac{\mathcal{Y}(\varepsilon_1 + \varepsilon)}{\varepsilon_1 - \tilde{E}} d\varepsilon_1 \int_0^D \frac{\mathcal{Y}(\varepsilon_2 + \varepsilon_1)}{\varepsilon_2 - \tilde{E}} d\varepsilon_2 \right] \end{aligned}$$

$$+ 5^3 \left(\frac{J\rho}{4N} \right)^6 \int_0^D \frac{y(\varepsilon_1 + \varepsilon)}{\varepsilon_1 - \tilde{E}} d\varepsilon_1 \int_0^D \frac{y(\varepsilon_2 + \varepsilon_1)}{\varepsilon_2 - \tilde{E}} d\varepsilon_2 \int_0^D \frac{y(\varepsilon_3 + \varepsilon_2)}{\varepsilon_3 - \tilde{E}} d\varepsilon_3 + \dots]. \quad (\text{A4})$$

$$y(\varepsilon + \varepsilon_1) = \log \left[\frac{\varepsilon + \varepsilon_1 - \tilde{E}}{\varepsilon + \varepsilon_1 + D - \tilde{E}} \right]. \quad (\text{A5})$$

Retaining the most divergent terms, we obtain the following power series with respect to $x = (J\rho/4N) \log(|\tilde{E}|/D)$:

$$1 = -x + \frac{5}{3}x^3 - \frac{10}{3}x^5 + \frac{425}{63}x^7 - \frac{7750}{567}x^9 + \dots. \quad (\text{A6})$$

The ratios between the coefficients of the two successive terms on the right-hand side of the power series are -1.667 , -2 , -2.024 and -2.026 . Therefore, the radius of convergence of this series is less than about $1/\sqrt{2} \simeq 0.70$ and within this radius Eq.(A6) has no root. This is also true on the minus side of x , which corresponds to the ferromagnetic exchange.

For $(J\rho/4N) = \alpha < 0$, it is expected that (A1.2.3) has no solution. This equation can be expressed as

$$\Gamma(x) = \frac{\alpha}{x + w} \int_0^1 \Gamma(x') [1 + 5\alpha \log(x + x' + w)] dx', \quad (\text{A7})$$

where $w = -\tilde{E}/D$. Then, the kernel, $[1 + 5\alpha \log(x + x' + w)]$, of this integral equation is always positive for negative α . Therefore, if $\Gamma(x)$ has no node, it is easy to see that (A7) has no solution. If $\Gamma(x)$ has one zero at $x = x_0$, the following relation should hold:

$$\left\{ \int_0^{x_0} \Gamma(x') [1 + 5\alpha \log(x + x' + w)] dx' + \int_{x_0}^1 \Gamma(x') [1 + 5\alpha \log(x + x' + w)] dx' \right\}_{x=x_0} = 0. \quad (\text{A8})$$

We assume that $\Gamma(x) > 0$ for $x < x_0$. When x is smaller than x_0 , the left-hand side of (A8) has the same sign as that of $\Gamma(x)$ for $x < x_0$, namely plus sign. However, the left-hand side of (A7) should have a sign opposite to (A8) because of negative α . This is impossible. The same reasoning can be applied to the cases for which $\Gamma(x)$ has any number of zeros, and we can verify that (A7) has no solution for negative α . Thus, it can be concluded that a bound state which appears in the perturbation series is not realistic.

References

- 1) K. Yosida, Phys. Rev. **147** (1966), 223.

- 2) A. Okiji, Prog. Theor. Phys. **36** (1966), 712.
- 3) K. Yosida, Prog. Theor. Phys. **36** (1966), 875.
- 4) H. Suhl and D. Wong, Physics **3** (1967), 17.
- 5) F. Takano and T. Ogawa, Prog. Theor. Phys. **35** (1966), 343.
- 6) L. Dworin, Phys. Rev. Letters **16** (1966), 1042.
- 7) D. R. Hamann, to be published.