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# Bound State due to the *s-d* Exchange Interaction

## ——*Effect of the Higher Order Perturbation*——

Ayao OKIJI

*Institute for Solid State Physics*
*University of Tokyo, Azabu, Tokyo*

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Higher order corrections to the binding energy of the singlet bound state obtained by Yosida for the case of an antiferromagnetic *s-d* exchange interaction are calculated. It is found that higher order terms up to the sixth order in $J$ increase the binding energy and consequently that the stability of singlet bound state is increased

## § 1. Introduction

The system considered in this paper consists of the conduction electrons which couple with a localized spin by the *s-d* exchange interaction. In this system it has been shown that when the interaction is antiferromagnetic, the usual perturbation expansion for physical quantities such as the scattering amplitude[1] and the magnitude of the localized spin[2] loses its meaning below a critical temperature. Many investigations have been made to explain the origin of these anomalies.[1),2)]

In view of this situation, Yosida has recently shown that a singlet bound state is realized for the case of an antiferromagnetic exchange interaction,[3] and concluded that the usual perturbation method breaks down for this reason. In his theory he starts with the state of a free electron gas in which one electron is excited above the Fermi sea and treats the effect of the *s-d* exchange interaction of the conduction electrons with a localized spin whose magnitude is one half by a generalized perturbation method. In the zeroth approximation of this theory the localized spin couples with an electron excited above the Fermi level by the *s-d* exchange interaction and makes a singlet bound state. In the first approximation which takes into account the effect of an excited electron-hole pair, this singlet bound state still survives for the case of antiferromagnetic interaction. From this calculation he concluded that the unphysical results obtained by the usual perturbation method which starts from the degenerate localized spin states originate in the fact that it disregards the existence of the singlet bound state.

However, it is not clear whether the singlet bound state obtained in the first approximation remains unchanged even when the approximation proceeds up to higher order. The purpose of this paper is to calculate the higher-order effects and to confirm the above conclusion obtained by Yosida.

## § 2. The method

The Hamiltonian of the system consisting of the conduction electrons and a localized spin situated at the origin is given by

$$H=\sum_{k\sigma}\varepsilon_{k\sigma}a_{k\sigma}^{*}a_{k\sigma}-\frac{J}{2N}\sum_{kk'}\{(a_{k'\uparrow}^{*}a_{k\uparrow}-a_{k'\downarrow}^{*}a_{k\downarrow})S_z+a_{k'\uparrow}^{*}a_{k\downarrow}S_{-}+a_{k'\downarrow}^{*}a_{k\uparrow}S_{+}\}, \quad (1)$$

where $a_{k\sigma}^*$ and $a_{k\sigma}$ are creation and annihilation operators of a conduction electron with wave vector $k$ and spin $\sigma$, $\varepsilon_k$ is its band energy measured from the Fermi energy, $S_z$ and $S_{\pm}$ are the components of the localized spin and $J$ is the coupling constant.

We expand the ground state wave function in the following series according to Yosida:

$$\begin{aligned}\psi=[&\sum_{k1}(\Gamma_{k1}^{\alpha}a_{k1\downarrow}^{*}\alpha+\Gamma_{k1}^{\beta}a_{k1\uparrow}^{*}\beta)\\
&+\sum_{k1k2k3}(\Gamma_{k1k2k3}^{\alpha\downarrow}a_{k1\downarrow}^{*}a_{k2\downarrow}^{*}a_{k3\downarrow}\alpha+\Gamma_{k1k2k3}^{\beta\uparrow}a_{k1\uparrow}^{*}a_{k2\uparrow}^{*}a_{k3\uparrow}\beta\\
&\qquad+\Gamma_{k1k2k3}^{\alpha\uparrow}a_{k1\uparrow}^{*}a_{k2\downarrow}^{*}a_{k3\uparrow}\alpha+\Gamma_{k1k2k3}^{\beta\downarrow}a_{k1\downarrow}^{*}a_{k2\uparrow}^{*}a_{k3\downarrow}\beta)\\
&+\sum_{k1k2k3k4k5}(\Gamma_{k1k2k3k4k5}^{\alpha\downarrow\downarrow}a_{k1\downarrow}^{*}a_{k2\downarrow}^{*}a_{k3\downarrow}^{*}a_{k4\downarrow}a_{k5\downarrow}\alpha\\
&\qquad+\Gamma_{k1k2k3k4k5}^{\alpha\downarrow\uparrow}a_{k1\downarrow}^{*}a_{k2\downarrow}^{*}a_{k3\uparrow}^{*}a_{k4\downarrow}a_{k5\uparrow}\alpha\\
&\qquad+\Gamma_{k1k2k3k4k5}^{\alpha\uparrow\uparrow}a_{k1\downarrow}^{*}a_{k2\uparrow}^{*}a_{k3\uparrow}^{*}a_{k4\uparrow}a_{k5\uparrow}\alpha\\
&\qquad+\Gamma_{k1k2k3k4k5}^{\beta\uparrow\uparrow}a_{k1\uparrow}^{*}a_{k2\uparrow}^{*}a_{k3\uparrow}^{*}a_{k4\uparrow}a_{k5\uparrow}\beta\\
&\qquad+\Gamma_{k1k2k3k4k5}^{\beta\uparrow\downarrow}a_{k1\uparrow}^{*}a_{k2\uparrow}^{*}a_{k3\downarrow}^{*}a_{k4\uparrow}a_{k5\downarrow}\beta\\
&\qquad+\Gamma_{k1k2k3k4k5}^{\beta\downarrow\downarrow}a_{k1\uparrow}^{*}a_{k2\downarrow}^{*}a_{k3\downarrow}^{*}a_{k4\downarrow}a_{k5\downarrow}\beta)\\
&+\cdots]\psi_v, \end{aligned} \quad (2)$$

where $\psi_v$ denotes the wave function for the Fermi sea and $\alpha$ and $\beta$ mean up and down spin states for the localized spin, respectively. Henceforth we represent the indexes $k_i$ by $i$.

Then, the Schrödinger equation can be written as follows:

$$\begin{aligned}[&\sum_{1}(\varepsilon_1-E)\{\Gamma_1^{\alpha}a_{1\downarrow}^{*}\alpha+\Gamma_1^{\beta}a_{1\uparrow}^{*}\beta\}\\
&+\sum_{123}(\varepsilon_1+\varepsilon_2-\varepsilon_3-E)\{\Gamma_{123}^{\alpha\downarrow}a_{1\downarrow}^{*}a_{2\downarrow}^{*}a_{3\downarrow}\alpha+\Gamma_{123}^{\beta\uparrow}a_{1\uparrow}^{*}a_{2\uparrow}^{*}a_{3\uparrow}\beta\\
&\qquad+\Gamma_{123}^{\alpha\uparrow}a_{1\uparrow}^{*}a_{2\downarrow}^{*}a_{3\uparrow}\alpha+\Gamma_{123}^{\beta\downarrow}a_{1\downarrow}^{*}a_{2\uparrow}^{*}a_{3\downarrow}\beta\}\\
&+\sum_{12345}(\varepsilon_1+\varepsilon_2+\varepsilon_3-\varepsilon_4-\varepsilon_5-E)\{\Gamma_{12345}^{\alpha\downarrow\downarrow}a_{1\downarrow}^{*}a_{2\downarrow}^{*}a_{3\downarrow}^{*}a_{4\downarrow}a_{5\downarrow}\alpha\\
&\qquad+\Gamma_{12345}^{\alpha\downarrow\uparrow}a_{1\downarrow}^{*}a_{2\downarrow}^{*}a_{3\uparrow}^{*}a_{4\downarrow}a_{5\uparrow}\alpha\end{aligned}$$

$$+\Gamma^{\alpha\uparrow\uparrow}_{12345}\, a^*_{1\downarrow}\, a^*_{2\uparrow}\, a^*_{3\uparrow}\, a_{4\uparrow}\, a_{5\uparrow}\, \alpha$$
$$+\Gamma^{\beta\uparrow\uparrow}_{12345}\, a^*_{1\uparrow}\, a^*_{2\uparrow}\, a^*_{3\uparrow}\, a_{4\uparrow}\, a_{5\uparrow}\, \beta$$
$$+\Gamma^{\beta\uparrow\downarrow}_{12345}\, a^*_{1\uparrow}\, a^*_{2\uparrow}\, a^*_{3\downarrow}\, a_{4\uparrow}\, a_{5\downarrow}\, \beta$$
$$+\Gamma^{\beta\downarrow\downarrow}_{12345}\, a^*_{1\uparrow}\, a^*_{2\downarrow}\, a^*_{3\downarrow}\, a_{4\downarrow}\, a_{5\downarrow}\, \beta\}$$

$$+\frac{J}{4N}\{\sum_{12}\Gamma_1{}^{\alpha}\, a^*_{2\downarrow}\, \alpha-2\sum_{12}\Gamma_1{}^{\beta}\, a^*_{2\downarrow}\, \alpha\}+\frac{J}{4N}\{\sum_{12}\Gamma_1{}^{\beta}\, a^*_{2\uparrow}\, \beta-2\sum_{12}\Gamma_1{}^{\alpha}\, a^*_{2\uparrow}\, \beta\}$$

$$-\frac{J}{4N}\sum_{123}\{\Gamma^{\alpha\downarrow}_{123}\, a^*_{2\downarrow}\, \alpha-\Gamma^{\alpha\downarrow}_{123}\, a^*_{1\downarrow}\, \alpha-\Gamma^{\alpha\uparrow}_{123}\, a^*_{2\downarrow}\, \alpha+2\Gamma^{\beta\downarrow}_{123}\, a^*_{1\downarrow}\, \alpha\}$$

$$-\frac{J}{4N}\sum_{123}\{\Gamma^{\beta\uparrow}_{123}\, a^*_{2\uparrow}\, \beta-\Gamma^{\beta\uparrow}_{123}\, a^*_{1\uparrow}\, \beta-\Gamma^{\beta\downarrow}_{123}\, a^*_{2\uparrow}\, \beta+2\Gamma^{\alpha\uparrow}_{123}\, a^*_{1\uparrow}\, \beta\}$$

$$-\frac{J}{4N}\sum_{123}\{\Gamma_1{}^{\alpha}\, a^*_{1\downarrow}\, (a^*_{2\uparrow}\, a_{3\uparrow}-a^*_{2\downarrow}\, a_{3\downarrow})\, \alpha+2\Gamma_1{}^{\beta}\, a^*_{1\uparrow}\, a^*_{2\downarrow}\, a_{3\uparrow}\, \alpha\}$$

$$-\frac{J}{4N}\sum_{123}\{-\Gamma_1{}^{\beta}\, a^*_{1\uparrow}\, (a^*_{2\uparrow}\, a_{3\uparrow}-a^*_{2\downarrow}\, a_{3\downarrow})\, \beta+2\Gamma_1{}^{\alpha}\, a^*_{1\downarrow}\, a^*_{2\uparrow}\, a_{3\downarrow}\, \beta\}$$

$$-\frac{J}{4N}\sum_{1234}\{-\Gamma^{\alpha\downarrow}_{123}\, a^*_{2\downarrow}\, a_{3\downarrow}\, a^*_{4\downarrow}\, \alpha+\Gamma^{\alpha\downarrow}_{123}\, a^*_{1\downarrow}\, a_{3\downarrow}\, a^*_{4\downarrow}\, \alpha+\Gamma^{\alpha\downarrow}_{123}\, a^*_{1\downarrow}\, a^*_{2\downarrow}\, a_{4\downarrow}\, \alpha$$
$$-2\Gamma^{\beta\downarrow}_{123}\, a^*_{1\downarrow}\, a_{3\downarrow}\, a^*_{4\downarrow}\, \alpha\}$$

$$-\frac{J}{4N}\sum_{1234}\{\Gamma^{\alpha\uparrow}_{123}\, a^*_{2\downarrow}\, a_{3\uparrow}\, a^*_{4\uparrow}\, \alpha-\Gamma^{\alpha\uparrow}_{123}\, a^*_{1\uparrow}\, a^*_{2\downarrow}\, a_{4\uparrow}\, \alpha+\Gamma^{\alpha\uparrow}_{123}\, a^*_{1\uparrow}\, a_{3\uparrow}\, a^*_{4\downarrow}\, \alpha$$
$$+2\Gamma^{\beta\uparrow}_{123}\, a^*_{2\uparrow}\, a_{3\uparrow}\, a^*_{4\downarrow}\, \alpha-2\Gamma^{\beta\uparrow}_{123}\, a^*_{1\uparrow}\, a_{3\uparrow}\, a^*_{4\downarrow}\, \alpha-2\Gamma^{\beta\downarrow}_{123}\, a^*_{1\downarrow}\, a^*_{2\uparrow}\, a_{4\uparrow}\, \alpha\}$$

$$-\frac{J}{4N}\sum_{1234}\{-\Gamma^{\beta\uparrow}_{123}\, a^*_{2\uparrow}\, a_{3\uparrow}\, a^*_{4\uparrow}\, \beta+\Gamma^{\beta\uparrow}_{123}\, a^*_{1\uparrow}\, a_{3\uparrow}\, a^*_{4\uparrow}\, \beta+\Gamma^{\beta\uparrow}_{123}\, a^*_{1\uparrow}\, a^*_{2\uparrow}\, a_{4\uparrow}\, \beta$$
$$-2\Gamma^{\alpha\uparrow}_{123}\, a^*_{1\uparrow}\, a_{3\uparrow}\, a^*_{4\uparrow}\, \beta\}$$

$$-\frac{J}{4N}\sum_{1234}\{\Gamma^{\beta\downarrow}_{123}\, a^*_{1\downarrow}\, a_{3\downarrow}\, a^*_{4\uparrow}\, \beta-\Gamma^{\beta\downarrow}_{123}\, a^*_{1\downarrow}\, a^*_{2\uparrow}\, a_{4\downarrow}\, \beta+\Gamma^{\beta\downarrow}_{123}\, a^*_{2\uparrow}\, a_{3\downarrow}\, a^*_{4\downarrow}\, \beta$$
$$+2\Gamma^{\alpha\downarrow}_{123}\, a^*_{2\downarrow}\, a_{3\downarrow}\, a^*_{4\uparrow}\, \beta-2\Gamma^{\alpha\downarrow}_{123}\, a^*_{1\downarrow}\, a_{3\downarrow}\, a^*_{4\uparrow}\, \beta-2\Gamma^{\alpha\uparrow}_{123}\, a^*_{1\uparrow}\, a^*_{2\downarrow}\, a_{4\downarrow}\, \beta\}$$

$$-\frac{J}{4N}\sum_{12345}\{\Gamma^{\alpha\downarrow\downarrow}_{12345}\, (a^*_{1\downarrow}\, a^*_{3\downarrow}\, a_{5\downarrow}+a^*_{1\downarrow}\, a^*_{2\downarrow}\, a_{4\downarrow}-a^*_{1\downarrow}\, a^*_{3\downarrow}\, a_{4\downarrow}-a^*_{1\downarrow}\, a^*_{2\downarrow}\, a_{5\downarrow}$$
$$+a^*_{2\downarrow}\, a^*_{3\downarrow}\, a_{4\downarrow}-a^*_{2\downarrow}\, a^*_{3\downarrow}\, a_{5\downarrow})\, \alpha-\Gamma^{\alpha\downarrow\uparrow}_{12345}\, a^*_{1\downarrow}\, a^*_{2\downarrow}\, a_{4\downarrow}\, \alpha$$
$$+2\Gamma^{\beta\downarrow\downarrow}_{12345}\, (a^*_{2\downarrow}\, a^*_{3\downarrow}\, a_{5\downarrow}-a^*_{2\downarrow}\, a^*_{3\downarrow}\, a_{4\downarrow})\, \alpha\}$$

$$-\frac{J}{4N}\sum_{12345}\{\Gamma^{\alpha\downarrow\uparrow}_{12345}\, (a^*_{1\downarrow}\, a^*_{3\uparrow}\, a_{5\uparrow}-a^*_{2\downarrow}\, a^*_{3\uparrow}\, a_{5\uparrow})\, \alpha$$
$$+\Gamma^{\alpha\uparrow\uparrow}_{12345}\, (a^*_{1\downarrow}\, a^*_{3\uparrow}\, a_{4\uparrow}+a^*_{1\downarrow}\, a^*_{2\uparrow}\, a_{5\uparrow}-a^*_{1\downarrow}\, a^*_{2\uparrow}\, a_{4\uparrow}-a^*_{1\downarrow}\, a^*_{3\uparrow}\, a_{5\uparrow})\, \alpha$$

$$+2\Gamma^{\beta\uparrow\downarrow}_{12345}(a^*_{1\uparrow}a^*_{3\downarrow}a_{4\uparrow}-a^*_{2\uparrow}a^*_{3\downarrow}a_{4\uparrow})\alpha\}$$

$$-\frac{J}{4N}\sum_{12345}\{\Gamma^{\beta\uparrow\uparrow}_{12345}(a^*_{1\uparrow}a^*_{3\uparrow}a_{5\uparrow}+a^*_{1\uparrow}a^*_{2\uparrow}a_{4\uparrow}-a^*_{1\uparrow}a^*_{2\uparrow}a_{5\uparrow}-a^*_{1\uparrow}a^*_{3\uparrow}a_{4\uparrow}$$

$$+a^*_{2\uparrow}a^*_{3\uparrow}a_{4\uparrow}-a^*_{2\uparrow}a^*_{3\uparrow}a_{5\uparrow})\beta-\Gamma^{\beta\uparrow\downarrow}_{12345}a^*_{1\uparrow}a^*_{2\uparrow}a_{4\uparrow}\beta$$

$$+2\Gamma^{\varkappa\uparrow\uparrow}_{12345}(a^*_{2\uparrow}a^*_{3\uparrow}a_{5\uparrow}-a^*_{2\uparrow}a^*_{3\uparrow}a_{4\uparrow})\beta\}$$

$$-\frac{J}{4N}\sum_{12345}\{\Gamma^{\beta\uparrow\downarrow}_{12345}(a^*_{1\uparrow}a^*_{3\downarrow}a_{5\downarrow}-a^*_{2\uparrow}a^*_{3\downarrow}a_{5\downarrow})\beta$$

$$+\Gamma^{\beta\downarrow\downarrow}_{12345}(a^*_{1\uparrow}a^*_{3\downarrow}a_{4\downarrow}+a^*_{1\uparrow}a^*_{2\downarrow}a_{5\downarrow}-a^*_{1\uparrow}a^*_{2\downarrow}a_{4\downarrow}-a^*_{1\uparrow}a^*_{3\downarrow}a_{5\downarrow})\beta$$

$$+2\Gamma^{\varkappa}_{12345}(a^*_{1\downarrow}a^*_{3\uparrow}a_{5\downarrow}-a^*_{2\downarrow}a^*_{3\uparrow}a_{4\downarrow})\beta\}$$

$$-\frac{J}{4N}\sum_{12345}\{\Gamma^{\varkappa\downarrow}_{123}a^*_{1\downarrow}a^*_{2\downarrow}a_{3\downarrow}(a^*_{4\uparrow}a_{5\uparrow}-a^*_{4\uparrow}a_{5\downarrow})\alpha$$

$$+\Gamma^{\varkappa\uparrow}_{123}a^*_{1\uparrow}a^*_{2\downarrow}a_{3\uparrow}(a^*_{4\uparrow}a_{5\uparrow}-a^*_{4\downarrow}a_{5\downarrow})\alpha$$

$$+2\Gamma^{\beta\uparrow}_{123}a^*_{1\uparrow}a^*_{2\uparrow}a_{3\uparrow}a^*_{4\downarrow}a_{5\uparrow}\alpha+2\Gamma^{\beta\downarrow}_{123}a^*_{1\downarrow}a^*_{2\uparrow}a_{3\downarrow}a^*_{4\downarrow}a_{5\uparrow}\alpha\}$$

$$-\frac{J}{4N}\sum_{12345}\{-\Gamma^{\beta\uparrow}_{123}a^*_{1\uparrow}a^*_{2\uparrow}a_{3\uparrow}(a^*_{4\uparrow}a_{5\uparrow}-a^*_{4\downarrow}a_{5\downarrow})\beta$$

$$-\Gamma^{\beta\downarrow}_{123}a^*_{1\downarrow}a^*_{2\uparrow}a_{3\downarrow}(a^*_{4\uparrow}a_{5\uparrow}-a^*_{4\downarrow}a_{5\downarrow})\beta$$

$$+2\Gamma^{\alpha\downarrow}_{123}a^*_{1\downarrow}a^*_{2\downarrow}a_{3\downarrow}a^*_{4\uparrow}a_{5\downarrow}\beta+2\Gamma^{\varkappa\uparrow}_{123}a^*_{1\uparrow}a^*_{2\downarrow}a_{3\uparrow}a^*_{4\uparrow}a_{5\downarrow}\beta\}$$

$$\cdots]\psi_v=0. \tag{3}$$

Here, the terms of the expansion needed up to the third approximation have been written down. From this expression, we can easily obtain the following simultaneous equations which determine the energy eigenvalue and $\Gamma_1$, $\Gamma_{123}\cdots$, etc.:

$$\Gamma_k{}^{\alpha}(\varepsilon_k-E)+\frac{J}{4N}\sum_1\Gamma_1{}^{\alpha}-\frac{J}{2N}\sum_1\Gamma_1{}^{\beta}-\frac{J}{4N}\sum_{12}(\Gamma^{\alpha\downarrow}_{[1k],2}-\Gamma^{\varkappa\uparrow}_{1k,2}+2\Gamma^{\beta\downarrow}_{k1,2})=0, \tag{4}$$

$$\Gamma_k{}^{\beta}(\varepsilon_k-E)+\frac{J}{4N}\sum_1\Gamma_1{}^{\beta}-\frac{J}{2N}\sum_1\Gamma_1{}^{\alpha}-\frac{J}{4N}\sum_{12}(\Gamma^{\beta\uparrow}_{[1k],2}-\Gamma^{\beta\downarrow}_{1k,2}+2\Gamma^{\varkappa\uparrow}_{k1,2})=0, \tag{5}$$

$$\Gamma^{\varkappa\downarrow}_{[12],3}(\varepsilon_1+\varepsilon_2-\varepsilon_3-E)+\frac{J}{4N}(\Gamma_1{}^{\alpha}-\Gamma_2{}^{\alpha})+\frac{J}{4N}\sum_4(\Gamma^{\alpha\downarrow}_{[42],3}+\Gamma^{\alpha\downarrow}_{[14],3}+\Gamma^{\varkappa\downarrow}_{[21],4}$$

$$+2\Gamma^{\beta\downarrow}_{24,3}-2\Gamma^{\beta\downarrow}_{14,3})+\frac{J}{4N}\sum_{45}(\Gamma^{\varkappa\downarrow\downarrow}_{[412],[53]}+\Gamma^{\varkappa\downarrow\uparrow}_{[12]4,35}+2\Gamma^{\beta\downarrow\downarrow}_{4[12],[35]})=0, \tag{6}$$

$$\Gamma^{\beta\uparrow}_{[12],3}(\varepsilon_1+\varepsilon_2-\varepsilon_3-E)+\frac{J}{4N}(\Gamma_1{}^{\beta}-\Gamma_2{}^{\beta})+\frac{J}{4N}\sum_4(\Gamma^{\beta\uparrow}_{[42],3}+\Gamma^{\beta\uparrow}_{[14],3}+\Gamma^{\beta\uparrow}_{[21],4}$$

$$+2\Gamma^{\varkappa\uparrow}_{24,3}-2\Gamma^{\varkappa\uparrow}_{14,3})+\frac{J}{4N}\sum_{45}(\Gamma^{\beta\uparrow\uparrow}_{[412],[53]}+\Gamma^{\beta\uparrow\downarrow}_{[12]4,35}+2\Gamma^{\varkappa\uparrow\uparrow}_{4[12],[35]})=0, \tag{7}$$

$$\Gamma^{\gamma\uparrow}_{12,3}(\varepsilon_1+\varepsilon_2-\varepsilon_3-E)+\frac{J}{4N}\Gamma_2{}^{\alpha}-\frac{J}{2N}\Gamma_1{}^{\beta}+\frac{J}{4N}\sum_4(\Gamma^{\gamma\uparrow}_{14,3}-\Gamma^{\gamma\uparrow}_{42,3}+\Gamma^{\gamma\uparrow}_{12,4}$$

$$+2\Gamma^{\beta\uparrow}_{[41],3}-2\Gamma^{\beta\downarrow}_{21,4})+\frac{J}{4N}\sum_{45}(\Gamma^{\gamma\downarrow\uparrow}_{[24]1,53}+\Gamma^{\gamma\uparrow\uparrow}_{2[14],[53]}+2\Gamma^{\beta\uparrow\downarrow}_{[41]2,35})=0, \tag{8}$$

$$\Gamma^{\beta\downarrow}_{12,3}(\varepsilon_1+\varepsilon_2-\varepsilon_3-E)+\frac{J}{4N}\Gamma_2{}^{\beta}-\frac{J}{2N}\Gamma_1{}^{\alpha}+\frac{J}{4N}\sum_4(\Gamma^{\beta\downarrow}_{11,3}-\Gamma^{\beta\downarrow}_{42,3}+\Gamma^{\beta\downarrow}_{12,4}$$

$$+2\Gamma^{\alpha\downarrow}_{[41],3}-2\Gamma^{\gamma\uparrow}_{21,4})+\frac{J}{4N}\sum_{45}(\Gamma^{\beta\uparrow\downarrow}_{[24]1,53}+\Gamma^{\beta\downarrow\downarrow}_{2[14],[53]}+2\Gamma^{\gamma\downarrow\uparrow}_{[41]2,35})=0, \tag{9}$$

$$\Gamma^{\gamma\downarrow\downarrow}_{[123],[45]}(\varepsilon_1+\varepsilon_2+\varepsilon_3-\varepsilon_4-\varepsilon_5-E)+\frac{J}{4N}(-\Gamma^{\gamma\downarrow}_{[12],4}+\Gamma^{\gamma\downarrow}_{[12],5}+\Gamma^{\gamma\downarrow}_{[13],4}-\Gamma^{\gamma\downarrow}_{[13],5}-\Gamma^{\gamma\downarrow}_{[23],4}$$

$$+\Gamma^{\gamma\downarrow}_{[23],5})+\frac{J}{4N}\sum_6(\Gamma^{\gamma\downarrow\downarrow}_{[623],[45]}+\Gamma^{\gamma\downarrow\downarrow}_{[163],[45]}+\Gamma^{\gamma\downarrow\downarrow}_{[126],[45]}-\Gamma^{\gamma\downarrow\downarrow}_{[123][65]}$$

$$-\Gamma^{\gamma\downarrow\downarrow}_{[123],[46]}-2\Gamma^{\beta\downarrow\downarrow}_{[623],[45]}+2\Gamma^{\beta\downarrow\downarrow}_{[613],[45]}-2\Gamma^{\beta\downarrow\downarrow}_{[612],[45]})+\cdots=0, \tag{10}$$

$$\Gamma^{\beta\uparrow\uparrow}_{[123],[45]}(\varepsilon_1+\varepsilon_2+\varepsilon_3-\varepsilon_4-\varepsilon_5-E)+\frac{J}{4N}(-\Gamma^{\beta\uparrow}_{[12],4}+\Gamma^{\beta\uparrow}_{[12],5}+\Gamma^{\beta\uparrow}_{[13],4}-\Gamma^{\beta\uparrow}_{[13],5}-\Gamma^{\beta\uparrow}_{[23],4}$$

$$+\Gamma^{\beta\uparrow}_{[23],5})+\frac{J}{4N}\sum_6(\Gamma^{\beta\uparrow\uparrow}_{[623],[45]}+\Gamma^{\beta\uparrow\uparrow}_{[163],[45]}+\Gamma^{\beta\uparrow\uparrow}_{[126],[45]}-\Gamma^{\beta\uparrow\uparrow}_{[123],[65]}$$

$$-\Gamma^{\beta\uparrow\uparrow}_{[123],[46]}-2\Gamma^{\gamma\uparrow\uparrow}_{[623],[45]}+2\Gamma^{\gamma\uparrow\uparrow}_{[613],[45]}-2\Gamma^{\gamma\uparrow\uparrow}_{[612],[45]})+\cdots=0, \tag{11}$$

$$\Gamma^{\gamma\downarrow\uparrow}_{[12]3,45}(\varepsilon_1+\varepsilon_2+\varepsilon_3-\varepsilon_4-\varepsilon_5-E)+\frac{J}{4N}(\Gamma^{\gamma\downarrow}_{[12],4}-\Gamma^{\gamma\uparrow}_{32,5}+\Gamma^{\gamma\uparrow}_{31,5}-2\Gamma^{\beta\downarrow}_{13,4}+2\Gamma^{\beta\downarrow}_{23,4})$$

$$+\frac{J}{4N}\sum_6(\Gamma^{\gamma\downarrow\uparrow}_{[62]3,45}+\Gamma^{\gamma\downarrow\uparrow}_{[16]3,45}-\Gamma^{\gamma\downarrow\uparrow}_{[12]6,45}-\Gamma^{\gamma\downarrow\uparrow}_{[12]3,65}+\Gamma^{\gamma\downarrow\uparrow}_{[12]3,46}$$

$$+2\Gamma^{\beta\uparrow\downarrow}_{[63]1,54}-2\Gamma^{\beta\uparrow\downarrow}_{[63]2,54}+2\Gamma^{\beta\downarrow\downarrow}_{3[12],[46]})+\cdots=0, \tag{12}$$

$$\Gamma^{\beta\uparrow\downarrow}_{[12]3,45}(\varepsilon_1+\varepsilon_2+\varepsilon_3-\varepsilon_4-\varepsilon_5-E)+\frac{J}{4N}(\Gamma^{\beta\uparrow}_{[12],4}-\Gamma^{\beta\downarrow}_{32,5}+\Gamma^{\beta\downarrow}_{31,5}-2\Gamma^{\gamma\uparrow}_{13,4}+2\Gamma^{\gamma\uparrow}_{23,4})$$

$$+\frac{J}{4N}\sum_6(\Gamma^{\beta\uparrow\downarrow}_{[62]3,45}+\Gamma^{\beta\uparrow\downarrow}_{[16]3,45}-\Gamma^{\beta\uparrow\downarrow}_{[12]6,45}-\Gamma^{\beta\uparrow\downarrow}_{[12]3,65}+\Gamma^{\beta\uparrow\downarrow}_{[12]3,46}$$

$$+2\Gamma^{\gamma\downarrow\uparrow}_{[63]1,54}-2\Gamma^{\gamma\downarrow\uparrow}_{[63]2,54}+2\Gamma^{\gamma\uparrow\uparrow}_{3[12],[46]})+\cdots=0, \tag{13}$$

$$\Gamma^{\gamma\uparrow\uparrow}_{1[23],[45]}(\varepsilon_1+\varepsilon_2+\varepsilon_3-\varepsilon_4-\varepsilon_5-E)+\frac{J}{4N}(-\Gamma^{\gamma\uparrow}_{21,4}+\Gamma^{\gamma\uparrow}_{21,5}+\Gamma^{\gamma\uparrow}_{31,4}-\Gamma^{\gamma\uparrow}_{31,5}+2\Gamma^{\beta\uparrow}_{[23],4}$$

$$-2\Gamma^{\beta\uparrow}_{[23],5})+\frac{J}{4N}\sum_6(\Gamma^{\gamma\uparrow\uparrow}_{6[23],[45]}-\Gamma^{\alpha\uparrow\uparrow}_{1[63],[45]}-\Gamma^{\alpha\uparrow\uparrow}_{1[26],[45]}+\Gamma^{\gamma\uparrow\uparrow}_{1[23],[65]}+\Gamma^{\gamma\uparrow\uparrow}_{1[23],[46]}$$

$$-2\Gamma^{\beta\uparrow\uparrow}_{[623],[45]}+2\Gamma^{\beta\uparrow\downarrow}_{[23]1,46}-2\Gamma^{\beta\uparrow\downarrow}_{[23]1,56})+\cdots=0, \tag{14}$$

$$\Gamma^{\beta\downarrow\downarrow}_{1[23],[45]}(\varepsilon_1+\varepsilon_2+\varepsilon_3-\varepsilon_4-\varepsilon_5-E)+\frac{J}{4N}(-\Gamma^{\beta\downarrow}_{21,4}+\Gamma^{\beta\downarrow}_{21,5}+\Gamma^{\beta\downarrow}_{31,4}-\Gamma^{\beta\downarrow}_{31,5}+2\Gamma^{\gamma\downarrow}_{[23],4}$$

$$-2\Gamma^{\alpha\downarrow}_{[23],5}) + \frac{J}{4N}\sum_{6}(\Gamma^{\beta\downarrow\downarrow}_{1[23],[45]} - \Gamma^{\beta\downarrow\downarrow}_{1[63],[45]} - \Gamma^{\beta\downarrow\downarrow}_{1[26],[45]} + \Gamma^{\beta\downarrow\downarrow}_{1[23],[65]} + \Gamma^{\beta\downarrow\downarrow}_{1[23],[46]}$$

$$-2\Gamma^{\alpha\downarrow\downarrow}_{[623],[45]} + 2\Gamma^{\alpha\downarrow\uparrow}_{[23]1,46} - 2\Gamma^{\alpha\downarrow\uparrow}_{[23]1,56}) + \cdots = 0. \tag{15}$$

In the above expressions, we use abbreviation symbols such that $\Gamma_{[12],3}$ means $\Gamma_{12,3} - \Gamma_{21,3}$. The suffixes attached to $\Gamma$, 1 and 2 on the left side of the comma and 3 on the right side of the comma represent, respectively, the wave vector of the electron and the hole.

## § 3. The solution to the secular equation

First we shall explain the process of solving the somewhat complicated secular equation given by the series Eqs. (4) to (15). For simplicity, we represent $\Gamma_{12\ n}$ or the sum of $\Gamma_{12\ n}$ by $\Gamma_n$ and $(\varepsilon_1 + \varepsilon_2 + \cdots - \varepsilon_n - E)$ by $(n)$. Then we can write down Eqs. (4) to (15) symbolically as follows:

$$(1) \quad \Gamma_1 = J\Gamma_1 + J\Gamma_3, \tag{16}$$

$$(3) \quad \Gamma_3 = J\Gamma_1 + J\Gamma_3 + J\Gamma_5, \tag{17}$$

$$(5) \quad \Gamma_5 = J\Gamma_3 + J\Gamma_5 + J\Gamma_7, \tag{18}$$

$$(7) \quad \Gamma_7 = \cdots \text{etc.}$$

Putting Eqs. (17), (18), etc., into Eq. (16) successively, we can eliminate $\Gamma_3$, $\Gamma_5$, etc., from Eq. (16) and obtain the following expression:

$$(1) \quad \Gamma_1 = J\Gamma_1 + J^2\frac{\Gamma_1}{(3)} + J^3\frac{\Gamma_1}{(3)(3)} + J^4\left(\frac{\Gamma_1}{(5)(3)(3)} + \frac{\Gamma_1}{(3)(3)(3)}\right)$$

$$+ J^5\left(\frac{\Gamma_1}{(5)(5)(3)(3)} + \frac{2\Gamma_1}{(5)(3)(3)(3)} + \frac{\Gamma_1}{(3)(3)(3)(3)}\right) + J^6(\cdots). \tag{19}$$

As is seen in Eq. (19), there are two kinds of processes in fourth order in $J$ and three kinds of processes in fifth order in $J$, etc. The calculations are carried out up to fifth order in $J$. However, in order to avoid lengthy expressions, we write down here only the expression up to fourth order in $J$:

$$\Gamma_k^{\alpha}(\varepsilon_k - E) = -\frac{J}{4N}\sum_{1}(\Gamma_1^{\alpha} - 2\Gamma_1^{\beta}) - \left(\frac{J}{4N}\right)^2\sum_{12}\frac{1}{(\varepsilon_k + \varepsilon_1 - \varepsilon_2 - E)}(\Gamma_1^{\alpha} + 4\Gamma_1^{\beta} - 6\Gamma_k^{\alpha})$$

$$-\left(\frac{J}{4N}\right)^3\sum_{123}\left[\frac{1}{(\varepsilon_k + \varepsilon_1 - \varepsilon_2 - E)}\left\{\frac{1}{(\varepsilon_k + \varepsilon_3 - \varepsilon_2 - E)}(-5\Gamma_3^{\alpha} - 2\Gamma_3^{\beta} + 12\Gamma_k^{\alpha})\right.\right.$$

$$+\frac{1}{(\varepsilon_1 + \varepsilon_3 - \varepsilon_2 - E)}(-2\Gamma_3^{\alpha} + 4\Gamma_3^{\beta} - 5\Gamma_1^{\alpha} - 2\Gamma_1^{\beta})$$

$$\left.\left.+\frac{1}{(\varepsilon_k + \varepsilon_1 - \varepsilon_3 - E)}(\Gamma_1^{\alpha} - 14\Gamma_1^{\beta} + 12\Gamma_k^{\alpha})\right\}\right]$$

$$
\begin{aligned}
-\left(\frac{J}{4N}\right)^4 \sum_{1234} \Bigg[ & \frac{1}{(\varepsilon_k+\varepsilon_1+\varepsilon_3-\varepsilon_2-\varepsilon_4-E)\,(\varepsilon_k+\varepsilon_1-\varepsilon_2-E)} \\
& \times \left\{ \frac{1}{(\varepsilon_k+\varepsilon_1-\varepsilon_2-E)} (-36\Gamma_k{}^\alpha+6\Gamma_1{}^\alpha+24\Gamma_1{}^\beta) \right. \\
& + \frac{1}{(\varepsilon_1+\varepsilon_3-\varepsilon_2-E)} (7\Gamma_1{}^\alpha-8\Gamma_1{}^\beta+22\Gamma_3{}^\alpha-8\Gamma_3{}^\beta) \\
& + \frac{1}{(\varepsilon_k+\varepsilon_3-\varepsilon_2-E)} (-6\Gamma_k{}^\alpha+7\Gamma_3{}^\alpha-8\Gamma_3{}^\beta) \\
& + \frac{1}{(\varepsilon_k+\varepsilon_1-\varepsilon_4-E)} (-6\Gamma_k{}^\alpha-5\Gamma_1{}^\alpha+16\Gamma_1{}^\beta) \\
& + \frac{1}{(\varepsilon_k+\varepsilon_3-\varepsilon_4-E)} (12\Gamma_k{}^\alpha-2\Gamma_3{}^\alpha-8\Gamma_3{}^\beta) \\
& \left. + \frac{1}{(\varepsilon_1+\varepsilon_3-\varepsilon_4-E)} (-2\Gamma_1{}^\alpha-8\Gamma_1{}^\beta+3\Gamma_3{}^\alpha) \right\} \\
& + \frac{1}{(\varepsilon_k+\varepsilon_1-\varepsilon_2-E)\,(\varepsilon_1+\varepsilon_3-\varepsilon_2-E)} \\
& \times \left\{ \frac{1}{(\varepsilon_1+\varepsilon_4-\varepsilon_2-E)} (-14\Gamma_4{}^\alpha-8\Gamma_4{}^\beta+13\Gamma_1{}^\alpha+16\Gamma_1{}^\beta) \right. \\
& + \frac{1}{(\varepsilon_3+\varepsilon_4-\varepsilon_2-E)} (-3\Gamma_4{}^\alpha+4\Gamma_3{}^\alpha-8\Gamma_3{}^\beta) \\
& \left. + \frac{1}{(\varepsilon_1+\varepsilon_3-\varepsilon_4-E)} (4\Gamma_3{}^\alpha-8\Gamma_3{}^\beta+7\Gamma_1{}^\alpha-8\Gamma_1{}^\beta) \right\} \\
& + \frac{1}{(\varepsilon_k+\varepsilon_1-\varepsilon_2-E)\,(\varepsilon_k+\varepsilon_3-\varepsilon_2-E)} \\
& \times \left\{ \frac{1}{(\varepsilon_3+\varepsilon_4-\varepsilon_2-E)} (4\Gamma_4{}^\alpha-8\Gamma_4{}^\beta+25\Gamma_3{}^\alpha-8\Gamma_3{}^\beta) \right. \\
& + \frac{1}{(\varepsilon_k+\varepsilon_4-\varepsilon_2-E)} (13\Gamma_4{}^\alpha+16\Gamma_4{}^\beta-42\Gamma_k{}^\alpha) \\
& \left. + \frac{1}{(\varepsilon_k+\varepsilon_3-\varepsilon_4-E)} (-6\Gamma_k{}^\alpha-5\Gamma_3{}^\alpha+16\Gamma_3{}^\beta) \right\} \\
& + \frac{1}{(\varepsilon_k+\varepsilon_1-\varepsilon_2-E)\,(\varepsilon_k+\varepsilon_1-\varepsilon_3-E)} \\
& \times \left\{ \frac{1}{(\varepsilon_1+\varepsilon_4-\varepsilon_3-E)} (4\Gamma_4{}^\alpha-8\Gamma_4{}^\beta-5\Gamma_1{}^\alpha+16\Gamma_1{}^\beta) \right.
\end{aligned}
$$

$$+\frac{1}{(\varepsilon_k+\varepsilon_4-\varepsilon_3-E)}(7\Gamma_4^\alpha-8\Gamma_4^\beta-6\Gamma_k^\alpha)$$

$$+\frac{1}{(\varepsilon_k+\varepsilon_1-\varepsilon_4-E)}(-42\Gamma_k^\alpha+\Gamma_1^\alpha+40\Gamma_1^\beta)\Big\}\Big] \tag{20}$$

The terms proportional to $\Gamma_k$ on the right-hand side of Eq. (20) can be regarded as the energy shift of the band energy $\varepsilon_k$ and this gives no essential effect on the result, as has been discussed in reference 3).

For the singlet state $(\Gamma_k^\alpha=-\Gamma_k^\beta)$, we write Eq. (20) in the form

$$\Gamma_k^\alpha=-3\frac{J}{4N}\frac{1}{(\varepsilon_k-E)}(G+f(\varepsilon_k)), \tag{21}$$

where

$$G=\sum_k \Gamma_k.$$

The higher order terms in $J$ are included in $f(\varepsilon)$. Then we insert Eq. (21) into Eq. (20). Neglecting the energy shift, we can write down Eq. (20) up to fourth order in $J$ as follows:

$$1=-3\frac{\rho J}{4N}\int_0^D\frac{1}{(\varepsilon_k-E)}d\varepsilon_k+3\left(\frac{\rho J}{4N}\right)^3\int_0^D\int_0^D\int_{-D}^0\frac{-3}{(\varepsilon_k-E)(\varepsilon_1-E)(\varepsilon_k+\varepsilon_1-\varepsilon_2-E)}d\varepsilon_k d\varepsilon_1 d\varepsilon_2$$

$$+3\left(\frac{\rho J}{4N}\right)^4\Big[\int_0^D\int_0^D\int_0^D\int_{-D}^0\frac{-3}{(\varepsilon_k-E)(\varepsilon_3-E)(\varepsilon_k+\varepsilon_1-\varepsilon_2-E)(\varepsilon_k+\varepsilon_3-\varepsilon_2-E)}d\varepsilon_k d\varepsilon_1 d\varepsilon_2 d\varepsilon_3$$

$$+\int_0^D\int_0^D\int_0^D\int_{-D}^0\frac{-6}{(\varepsilon_k-E)(\varepsilon_3-E)(\varepsilon_k+\varepsilon_1-\varepsilon_2-E)(\varepsilon_1+\varepsilon_3-\varepsilon_2-E)}d\varepsilon_k d\varepsilon_1 d\varepsilon_2 d\varepsilon_3$$

$$+\int_0^D\int_0^D\int_0^D\int_{-D}^0\frac{-3}{(\varepsilon_k-E)(\varepsilon_1-E)(\varepsilon_k+\varepsilon_1-\varepsilon_2-E)(\varepsilon_1+\varepsilon_3-\varepsilon_2-E)}d\varepsilon_k d\varepsilon_1 d\varepsilon_2 d\varepsilon_3$$

$$+\int_0^D\int_0^D\int_{-D}^0\int_{-D}^0\frac{15}{(\varepsilon_k-E)(\varepsilon_1-E)(\varepsilon_k+\varepsilon_1-\varepsilon_2-E)(\varepsilon_k+\varepsilon_1-\varepsilon_3-E)}d\varepsilon_k d\varepsilon_1 d\varepsilon_2 d\varepsilon_3\Big]. \tag{22}$$

There is no contribution to Eq. (22) from $f(\varepsilon)$ in Eq. (21) up to fourth order in $J$ but the contribution of $f(\varepsilon)$ (i.e. the contribution of the second iteration) appears at the fifth order in $J$. Here we assume a constant state density $\rho$, and take $2D$ as the band width. Considering the above equation up to third order in $J$, we obtain Yosida's result.[3] The integrals for the fourth order in $J$ are evaluated in the Appendix. In Eq. (22) the second integral of the fourth order in $J$ gives a logarithmic term of lower order and a regular term. Thus, by retaining the logarithmic terms of the highest order, Eq. (22) is reduced to

$$1=3x+3x^3+6x^4,$$

where $x$ means $(\rho J/4N)\log|E/D|$. The solution to this equation is

$$x_0=0.295.$$

The calculation for the fifth order in $J$ becomes somewhat complicated but is similar to that for the fourth order in $J$. In this order there appear many terms which do not contribute to the logarithmic terms in the highest order. The result reduces to

$$+\left(\frac{3}{5}+\frac{69}{5}\right)x^5.$$

The first and second terms in the above expression correspond to $\Gamma_1/(5)(3)(3)$ and $\Gamma_1/(3)(3)(3)$ in the symbolical equation (19), respectively. It should be noted here that there is another contribution to this order in $J$, which arises from the second iteration. This contribution which has been calculated by Yosida (see Eq. (31) of reference 3)), amounts to $+(18/5)x^5$. Thus the total contribution to the terms of fifth order in $J$ is obtained as $+18x^5$. Taking into account this term, we obtain the root of the polynomial with respect to $x$, $x_0$, as follows:

$$x_0=0.286.$$

For the terms of sixth order in $J$ the calculations become complicated but in principle they are similar to those of lower order in $J$. Therefore, we show only the final result,

$$+\left(\frac{4}{5}+\frac{20}{5}+\frac{168}{5}+\frac{78}{5}\right)x^6=+54x^6.$$

The first, second and third terms in this expression correspond to $\Gamma_1/(5)(5)(3)(3)$, $\Gamma_1/(5)(3)(3)(3)$ and $\Gamma_1/(3)(3)(3)(3)$ in Eq. (19), respectively and the last term comes from the second iteration in Eq. (20).

Thus, the secular equation for the singlet state up to sixth order in $J$ becomes

$$1=3x+3x^3+6x^4+18x^5+54x^6, \tag{23}$$

the solution to which is obtained as

$$x_0=0.280 \qquad \text{or} \qquad \frac{\rho J}{N}\log\left|\frac{E}{D}\right|=1.12. \tag{24}$$

It should be noted that since each coefficient of $x^n$ has a plus sign, the value of $x_0$ decreases as one goes to higher approximations. This means that the singlet bound state becomes more stable as a result of the higher order corrections.

Therefore, we may consider that the excited electron-hole pairs give an effect favorable to the singlet bound state which has been obtained in the zero-approximation.

For the case of a ferromagnetic interaction ($\Gamma_k^\alpha = \Gamma_k^\beta$), the calculations are similar to those for the antiferromagnetic interaction. Omitting details of the calculations, we give the final result below.

$$1 = -x + \frac{5}{3}x^3 + \frac{10}{3}x^4 + \frac{14}{3}x^5 + \frac{62}{9}x^6. \tag{25}$$

In the zeroth approximation, the solution $x_0 = -1$ is obtained. However, up to $x^3$ there is no solution for negative values of $x$.[3] This situation is repeated up to $x^6$. That is, if we take into account the $x^4$ term, there is a solution for negative values of $x$ but there is no solution when we take into account the $x^5$ term. In this situation it is natural to conclude that there is no bound state for the case of a ferromagnetic interaction.

In the above calculations we omitted the self-energy parts. These are the terms proportional to $\Gamma_k$ on the right-hand side of Eq. (20). They can be written up to third order in $J$ as

$$+\left(\frac{J}{4N}\right)^2 \sum_{12} \frac{6}{(\varepsilon_k+\varepsilon_1-\varepsilon_2-E)}\Gamma_k^\alpha$$

$$+\left(\frac{J}{4N}\right)^3 \sum_{123} \left\{\frac{-12}{(\varepsilon_k+\varepsilon_1-\varepsilon_2-E)(\varepsilon_k+\varepsilon_3-\varepsilon_2-E)} + \frac{-12}{(\varepsilon_k+\varepsilon_1-\varepsilon_2-E)(\varepsilon_k+\varepsilon_1-\varepsilon_3-E)}\right\}\Gamma_k^\alpha$$

$$+\cdots.$$

As can be easily seen, the parts, $\Delta E$, which are independent of $\varepsilon_k$ and $E$, namely the expression obtained by putting $\varepsilon_k = 0$ and $E = 0$ in the above expression, can be included in $E$ on the left-hand side of Eq. (20). (The parts dependent on $\varepsilon_k$ may be renormalizable in $\varepsilon_k$ itself.) The expressions for $\Delta E$ are obviously equal to the free energy shift obtained by the usual perturbation method for the normal state. Similarly, we can show that the expression for $\Delta E$ obtained for the fourth order term in $J$ is the same as that for the fourth order energy shift calculated by the normal perturbation approach,[4] as far as the energy shift which should be included in $E$ on the left-hand side of Eq. (20) is concerned.

Finally we add one result obtained for the case of a localized spin $S$ which is greater than one half, although we do not enter into details. The calculation for the case in which one conduction electron spin is trapped by the localized spin $S$, can be carried out by a simple extension of the present calculation. The result obtained up to the fifth order in $J$ for the case of antiferromagnetic interaction is

$$1 = (S+1)2x - 1/3(S+1)(S^2-1)(2x)^3 - 1/3(S+1)(S^2-1)(2x)^4$$
$$+(1/30)(S+1)(2S^2-3)(2S^2-5)(2x)^5.$$

In the limiting case where $S\to\infty$, $J\to 0$ and $J\cdot S=$constant, the equation becomes

$$1=2Sx-(1/3)(2Sx)^3+(2/15)(2Sx)^5-\cdots.$$

If we notice that the right-hand side of this expression coincides with the expansion of $\tanh(2Sx)$ with respect to $2Sx$, it may be written

$$1=\tanh(2Sx).$$

Therefore in this limiting case no bound state appears, as we should expect. This result can be considered as a partial justification of our treatment of the present problem.

## § 4. Summary and conclusion

As the zeroth approximation, we have considered the state in which an electron excited above the Fermi sea couples with a localized spin (of magnitude one half) by the *s-d* exchange interaction. Then, we take into account the effect of excited electron-hole pairs by a generalized perturbation method. These calculations have been carried out up to the fourth approximation. The results are as follows:

1) For the case of an antiferromagnetic exchange interaction there appears a singlet bound state which is formed by the localized spin and the conduction electrons. Its binding energy is given by $D\exp(-1.12\cdot N/\rho|J|)$ in the highest approximation made in this paper.

2) For the case of a ferromagnetic interaction, the bound state which appeared in the zeroth approximation vanishes in approximations up to odd powers of $x$ but reappears in approximations up to even powers of $x$. In this sense, the bound state is unstable and it seems to be plausible that there is no bound state for the ferromagnetic interaction.

The results obtained here strongly support the conclusion derived by Yosida that for the case of antiferromagnetic interaction the ground state of the system is a singlet bound state and that the unphysical results obtained by the usual perturbation method which starts from degenerate localized spin states originate in the fact that it disregards the existence of the singlet bound state.

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## Appendix

For the fourth order in $J$, we must calculate the following three kinds of integrals. The first is

$$\int_0^D\int_0^D\int_{-D}^0\int_{-D}^0 \frac{1}{(\varepsilon_k-E)(\varepsilon_1-E)(\varepsilon_k+\varepsilon_1-\varepsilon_2-E)(\varepsilon_k+\varepsilon_1-\varepsilon_3-E)} d\varepsilon_k d\varepsilon_1 d\varepsilon_2 d\varepsilon_3 . \tag{A1}$$

In the above integral we can easily integrate the factors $(\varepsilon_k+\varepsilon_1-\varepsilon_2-E)^{-1}$ and $(\varepsilon_k+\varepsilon_1-\varepsilon_3-E)^{-1}$ with respect to $\varepsilon_2$ and $\varepsilon_3$:

$$\int_{-D}^0 \frac{1}{(\varepsilon_k+\varepsilon_1-\varepsilon_2-E)} d\varepsilon_2 = -\log\left|\frac{\varepsilon_k+\varepsilon_1-E}{\varepsilon_k+\varepsilon_1+D-E}\right| . \tag{A2}$$

This can be approximated by

$$-\log\left|\frac{\varepsilon_k+\varepsilon_1-E}{D}\right| . \tag{A3}$$

The difference between (A2) and (A3) gives logarithmic terms of lower order and regular terms in the result. Thus, the essential part of (A1) becomes

$$\int_0^D\int_0^D \frac{1}{(\varepsilon_k-E)(\varepsilon_1-E)} \log^2\left|\frac{\varepsilon_k+\varepsilon_1-E}{D}\right| d\varepsilon_k d\varepsilon_1 . \tag{A4}$$

In the next step we write the integrand as follows:

$$\frac{1}{(\varepsilon_k-E)(\varepsilon_1-E)}\log^2\left|\frac{\varepsilon_k+\varepsilon_1-E}{D}\right| = \frac{2}{(\varepsilon_k-E)(\varepsilon_k+\varepsilon_1-2E)}\log^2\left|\frac{\varepsilon_k+\varepsilon_1-E}{D}\right|$$

$$= \left\{\frac{2}{(\varepsilon_k-E)(\varepsilon_k+\varepsilon_1-E)} + \frac{2}{(\varepsilon_k-E)}\sum_{n=1}\frac{E^n}{(\varepsilon_k+\varepsilon_1-E)^{n+1}}\right\}\log^2\left|\frac{\varepsilon_k+\varepsilon_1-E}{D}\right| .$$

Neglecting the second term in the curly brackets for the same reason as stated above, we can integrate (A4) as

$$-\int_0^D \frac{2}{3}\frac{1}{(\varepsilon_k-E)}\log^3\left|\frac{\varepsilon_k-E}{D}\right| d\varepsilon_k .$$

Thus, we get $\frac{1}{6}\log^4|E/D|$ as the essential term. Here we have used the fact that $E$ is negative.

The second integral is calculated in a similar way:

$$\int_0^D\int_0^D\int_0^D\int_{-D}^0 \frac{1}{(\varepsilon_k-E)(\varepsilon_3-E)(\varepsilon_k+\varepsilon_1-\varepsilon_2-E)(\varepsilon_k+\varepsilon_3-\varepsilon_2-E)} d\varepsilon_k d\varepsilon_1 d\varepsilon_2 d\varepsilon_3$$

$$\cong -\int_0^D\int_0^D\int_{-D}^0 \frac{1}{(\varepsilon_k-E)(\varepsilon_3-E)(\varepsilon_k+\varepsilon_3-\varepsilon_2-E)}\log\left|\frac{\varepsilon_k-\varepsilon_2-E}{D}\right| d\varepsilon_k d\varepsilon_2 d\varepsilon_3$$

$$\cong \int_0^D\int_0^D \frac{1}{2}\frac{1}{(\varepsilon_k-E)(\varepsilon_3-E)}\log^2\left|\frac{\varepsilon_k+\varepsilon_3-E}{D}\right| d\varepsilon_k d\varepsilon_3 \cong \frac{1}{12}\log^4\left|\frac{E}{D}\right| .$$

The last one is

$$\int_0^D\int_0^D\int_0^D\int_{-D}^0 \frac{1}{(\varepsilon_k-E)(\varepsilon_3-E)(\varepsilon_k+\varepsilon_1-\varepsilon_2-E)(\varepsilon_1+\varepsilon_3-\varepsilon_2-E)} d\varepsilon_k d\varepsilon_1 d\varepsilon_2 d\varepsilon_3 .$$

This integral has no $\log^4|E/D|$ term.

For the fifth order in $J$, there are many types of integral. We choose the following two integrals as examples:

$$\int_0^D\int_0^D\int_0^D\int_{-D}^0\int_{-D}^0 \frac{1}{(\varepsilon_k-E)(\varepsilon_1-E)(\varepsilon_k+\varepsilon_1-\varepsilon_2-E)(\varepsilon_k+\varepsilon_1-\varepsilon_4-E)(\varepsilon_k+\varepsilon_1+\varepsilon_3-\varepsilon_2-\varepsilon_4-E)}$$
$$\times d\varepsilon_k d\varepsilon_1 d\varepsilon_2 d\varepsilon_3 d\varepsilon_4$$
$$\cong -\int_0^D\int_0^D\int_{-D}^0\int_{-D}^0 \frac{1}{(\varepsilon_k-E)(\varepsilon_1-E)(\varepsilon_k+\varepsilon_1-\varepsilon_2-E)(\varepsilon_k+\varepsilon_1-\varepsilon_4-E)}$$
$$\times \log\left|\frac{\varepsilon_k+\varepsilon_1-\varepsilon_2-\varepsilon_4-E}{D}\right| d\varepsilon_k d\varepsilon_1 d\varepsilon_2 d\varepsilon_4$$
$$\cong -\int_0^D\int_0^D\int_{-D}^0 \frac{1}{(\varepsilon_k-E)(\varepsilon_1-E)(\varepsilon_k+\varepsilon_1-\varepsilon_4-E)} \left\{\frac{1}{2}\log^2\left|\frac{\varepsilon_k+\varepsilon_1-\varepsilon_4-E}{D}\right|\right.$$
$$\left. -\log\left|\frac{\varepsilon_k+\varepsilon_1-E}{D}\right| \log\left|\frac{\varepsilon_k+\varepsilon_1-\varepsilon_4-E}{D}\right|\right\} d\varepsilon_k d\varepsilon_1 d\varepsilon_4$$
$$\cong -\int_0^D\int_0^D \frac{1}{3}\frac{1}{(\varepsilon_k-E)(\varepsilon_1-E)}\log^3\left|\frac{\varepsilon_k+\varepsilon_1-E}{D}\right| d\varepsilon_k d\varepsilon_1 = -\frac{1}{30}\log^5\left|\frac{E}{D}\right| ,$$

$$\int_0^D\int_0^D\int_0^D\int_{-D}^0\int_{-D}^0 \frac{1}{(\varepsilon_k-E)(\varepsilon_3-E)(\varepsilon_k+\varepsilon_1-\varepsilon_2-E)(\varepsilon_k+\varepsilon_3-\varepsilon_2-E)(\varepsilon_k+\varepsilon_3-\varepsilon_4-E)}$$
$$\times d\varepsilon_k\, d\varepsilon_1\, d\varepsilon_2\, d\varepsilon_3\, d\varepsilon_4$$
$$\cong \int_0^D\int_0^D\int_{-D}^0 \frac{1}{(\varepsilon_k-E)(\varepsilon_3-E)(\varepsilon_k+\varepsilon_3-\varepsilon_2-E)} \log\left|\frac{\varepsilon_k-\varepsilon_2-E}{D}\right| \log\left|\frac{\varepsilon_k+\varepsilon_3-E}{D}\right|$$
$$\times d\varepsilon_k\, d\varepsilon_2\, d\varepsilon_3$$
$$\cong -\int_0^D\int_0^D \frac{1}{2}\frac{1}{(\varepsilon_k-E)(\varepsilon_3-E)}\log^3\left|\frac{\varepsilon_k+\varepsilon_3-E}{D}\right| d\varepsilon_k d\varepsilon_3 \cong -\frac{1}{20}\log^5\left|\frac{E}{D}\right| .$$

Other terms of this order and sixth-order terms can be calculated in a similar way but the calculations are somewhat complicated.

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