

Electron-spin polarization in EuS

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Abstract

The conduction-electron state in a ferromagnetic semiconductor is studied theoretically by applying the coherent-potential approximation to the s - f model. Using the mean-field theory for fluctuating f spins, the variation of the density of states with temperature is investigated for various values of IS/W . Here IS is the exchange interaction energy and W is the bandwidth of the conduction band. Temperature dependence of the electron-spin polarization, experimentally observed in EuS, can be explained satisfactorily within this approximation.

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1. Introduction

The s - f model is currently accepted as a basis for studying the conduction-electron state in ordinary magnetic semiconductors such as Eu-chalcogenides^{1,2)}. In this model, the total Hamiltonian, H_t , consists of H_s , H_f and H_{sf} which represent the translational energy of an s electron, the Heisenberg-type exchange interaction between f spins, and the s - f exchange interaction between an s electron and f spins, respectively.

$$H_t = H_s + H_f + H_{sf} \quad (1.1)$$

$$H_s = \sum_{k\mu} \epsilon_k a_{k\mu}^\dagger a_{k\mu} \quad (1.2)$$

$$H_f = - \sum_{mn} J_{mn} S_m \cdot S_n \quad (1.3)$$

$$H_{sf} = -I \sum_{m\mu\nu} a_{m\mu}^\dagger \boldsymbol{\sigma} \cdot S_m a_{m\nu} \quad (1.4)$$

The notations used here are the same as in the previous papers³⁻⁵⁾.

A single electron (hereafter referred to as s electron), injected into an otherwise empty conduction band, moves in the crystal, while interacting with f spins through the s - f exchange interaction. Thus the conduction (s -) electron state in a magnetic semiconductor is strongly affected by the magnetic order of the f spins.

In the case of a ferromagnetic semiconductor, the orientations of f spins are completely random at the high-temperature limit ($T=\infty$), while as the temperature decreases to the Curie temperature (T_c), the correlation between f spins becomes so strong that a short-range order is formed. At even lower temperature, $T < T_c$, spontaneous magnetic (f spin) ordering develops, and at $T=0$ the orientations of f spins are perfectly arranged in one direction (z direction).

In the previous papers^{3,4)}, we studied the variation of the density of states with temperature at all range of paramagnetic temperatures, taking account of the scattering due to f spin correlation together with multiple scattering on one site.

In this work, we study the conduction-electron state in a ferromagnetic semiconductor at all temperatures. However, it is too difficult to treat the scattering due to f spin correlation together with multiple scattering, because spontaneous magnetization arises below T_c . Here, we focus on multiple scattering of an s electron in a ferromagnetic semiconductor within the single-site approximation, ignoring f spin correlation⁵⁾.

Pioneering work on the single-site approximation in the ferromagnetic temperature

region was done by Kubo⁶⁾, who formulated the coherent potential approximation (CPA) in terms of an effective locator. A thermal average over the fluctuating f spin states is taken in the treatment using the Green's function technique, and the results seem reasonable. However, detailed comparison between the calculated results and the phenomena (optical and transport properties) observed in real magnetic semiconductors has not yet been performed. Furthermore, Kubo's method is not convenient for incorporating the exchange scattering due to the f spin correlation.

The CPA for the s - f model was also studied by Nolting and coworkers⁷⁻¹¹⁾. Starting from the atomic limit solution, they first presented the quasi-particle multiband picture for the conduction band in magnetic semiconductors⁷⁻⁹⁾. Using the quasi-particle levels and spectral weights calculated based on their concept, they formulated the CPA using an alloy analogy^{7,10,11)}. Their treatment, however, is somewhat questionable¹⁾, because the quasi-particle concept is fully realized only in the weak coupling region, as they acknowledged in their study¹²⁾. Furthermore, their results do not agree with Kubo's. This discrepancy results probably from the difference in the thermal-average operation concerning the fluctuation of f spin, as is discussed later.

The aim of this work is to present the CPA for the s - f model in a t matrix formula, and to investigate the conduction-electron states in ferromagnetic semiconductors within the single-site approximation.

The construction of this paper is as follows. In Sec. 2, we first derive the t matrix elements of the s - f exchange interaction for a single f spin embedded in the effective medium, and present the CPA condition using the t matrix elements for the ferromagnetic temperature region. In Sec. 3, the numerically calculated results for the density of states and the electron-spin polarization are shown as a function of the normalized temperature T/T_c and the ratio IS/W , and are compared with experimental results. In Sec. 4, the concluding remarks are presented.

2. Formalism

2.1. Basic considerations

In ordinary magnetic semiconductors, the magnetic excitation energy is very small compared with both the bandwidth W and the s - f exchange energy IS ; ^{1,2)} thus, the f spins are treated as a quasistatic system, or the thermal average for the fluctuating f spin is calculated at the last stage of the derivation of physical quantities. We further assume that the s electron does not polarize the f spins, although the s electron state is strongly affected by the state of f spins.

Thus, we define the single-electron Green's function³⁻⁵⁾,

$$G(\omega) = \frac{1}{\omega - H}, \quad (2.1)$$

with

$$H = H_s + H_{sf}, \quad (2.2)$$

and write its thermal average for f spin as $\langle G \rangle_{\text{av}}$.

In order to apply the multiple-scattering theory¹³⁾, we divide H into the unperturbed Hamiltonian K and the perturbation term V . When magnetization arises, an s electron in

a ferromagnetic semiconductor is subject to different effective potentials through the s - f exchange interaction according to the orientation of its spin. Thus, let us assume as effective medium where an s electron is subject to complex potential, Σ_\uparrow or Σ_\downarrow , according to the orientation of its spin. Then, an s electron moving in this effective medium is described by the (unperturbed) reference Hamiltonian K :

$$K = \sum_{k\mu} (\epsilon_k + \Sigma_\mu) a_{k\mu}^\dagger a_{k\mu}. \quad (2.3)$$

Thus, the perturbation term $V (= H - K)$ is written as a sum over each lattice site:

$$V = \sum_m v_m \quad (2.4)$$

with

$$v_m = \sum_{\mu\nu} a_{m\nu}^\dagger (-I\sigma \cdot S_m - \Sigma_\mu \delta_{\mu\nu}) a_{m\nu} \quad (2.5)$$

Next, using the reference Green's function P given by

$$P(\omega) = \frac{1}{\omega - K}, \quad (2.6)$$

we define the t matrix of the s - f exchange interaction as

$$t_m = v_m [1 - P v_m]^{-1}. \quad (2.7)$$

Note that K , and thus P , includes no f spin operator, and that t_m represents the complete scattering associated with the isolated potential v_m in the effective medium.

According to the multiple-scattering theory¹³⁾, the total scattering operator T , which is related to G as

$$G = P + PTP, \quad (2.8)$$

is expressed as the multiple-scattering series,

$$T = \sum_m t_m + \sum_m t_m P_n \sum_{n(\neq m)} t_n + \sum_m t_m P_n \sum_{n(\neq m)} t_n P_l \sum_{l(\neq n)} t_l + \cdots. \quad (2.9)$$

Within the single-site approximation, the condition

$$\langle t_m \rangle_{av} = 0 \quad \text{for any } m \quad (2.10)$$

leads to $\langle T \rangle_{av} = 0$ and thus $\langle G \rangle_{av} = P$. This is the CPA.

In this approximation, the density of states for the electron's spin $\mu (= \uparrow \text{ or } \downarrow)$, $D_\mu(\omega)$, is calculated using

$$D_\mu(\omega) = -\frac{1}{\pi} \text{Im} F_\mu(\omega) \quad (2.11)$$

Here $F_\mu(\omega) = \langle m\mu | P | m\mu \rangle$ (independent of m) is the diagonal moment of P in the Wannier representation, and is obtained from

$$F_\mu(\omega) = \frac{1}{N} \sum_k \langle k\mu | P | k\mu \rangle = \frac{1}{N} \sum_k \frac{1}{\omega - \epsilon_k - \Sigma_\mu}. \quad (2.12)$$

2.2 t matrix elements of s - f exchange interaction

In this subsection we show the explicit expression for the elements of the t matrix defined by Eq. (2.7): the site index m will be omitted in the operator to avoid confusion.

In order to show the resulting expression, it is convenient to introduce the following symbols:

$$V_{\uparrow} = -IS_z - \Sigma_{\uparrow}, \quad (2.13)$$

$$V_{\downarrow} = +IS_z - \Sigma_{\downarrow}, \quad (2.14)$$

$$U_{\uparrow} = -I(S_z - 1) - \Sigma_{\uparrow}, \quad (2.15)$$

$$U_{\downarrow} = +I(S_z + 1) - \Sigma_{\downarrow}, \quad (2.16)$$

$$W_{\uparrow} = I^2 S_- S_+ = I^2 [S(S+1) - S_z^2 - S_z], \quad (2.17)$$

$$W_{\downarrow} = I^2 S_+ S_- = I^2 [S(S+1) - S_z^2 + S_z]. \quad (2.18)$$

Here, S_z is the z -component of f spin: $S_+ = S_x + iS_y$ and $S_- = S_x - iS_y$. The physical meaning of the above symbols can be easily explained. $V_{\uparrow}(V_{\downarrow})$ is the spin-diagonal component of the s - f exchange interaction, wherein an s electron with $\uparrow(\downarrow)$ spin interacts with f spin located in the medium of $\Sigma_{\uparrow}(\Sigma_{\downarrow})$. $U_{\uparrow}(U_{\downarrow})$ is the spin-diagonal component of the s - f exchange interaction wherein an s electron with $\uparrow(\downarrow)$ spin interacts with f spin which has already flipped in the previous scattering; thus, the f spin operator S_z is replaced by $S_z - 1$ ($S_z + 1$). Both $V_{\uparrow}(V_{\downarrow})$ and $U_{\uparrow}(U_{\downarrow})$ describe the scattering process without spin-flip. On the other hand, $W_{\uparrow}(W_{\downarrow})$ is the interaction energy required by an s electron of $\uparrow(\downarrow)$ spin to flip and then reverse its spin after the intermediate propagation with flipped spin. It is worth noting that all the symbols defined above can be described in terms of S_z only.

We do not present tedious details, since the calculation is straight-forward and no more approximation is needed. The resulting expressions are

$$t_{\uparrow\uparrow} = \frac{V_{\uparrow} + F_{\downarrow}(W_{\uparrow} - V_{\uparrow}U_{\downarrow})}{1 - F_{\downarrow}U_{\downarrow} - F_{\uparrow}[V_{\uparrow} + F_{\downarrow}(W_{\uparrow} - V_{\uparrow}U_{\downarrow})]}, \quad (2.19)$$

$$t_{\downarrow\downarrow} = \frac{V_{\downarrow} + F_{\uparrow}(W_{\downarrow} - V_{\downarrow}U_{\uparrow})}{1 - F_{\uparrow}U_{\uparrow} - F_{\downarrow}[V_{\downarrow} + F_{\uparrow}(W_{\downarrow} - V_{\downarrow}U_{\uparrow})]}, \quad (2.20)$$

$$t_{\uparrow\downarrow} = \frac{1}{1 - F_{\downarrow}U_{\downarrow} - F_{\uparrow}[V_{\uparrow} + F_{\downarrow}(W_{\uparrow} - V_{\uparrow}U_{\downarrow})]} (-IS_-) \quad (2.21)$$

$$= (-IS_-) \frac{1}{1 - F_{\uparrow}U_{\uparrow} - F_{\downarrow}[V_{\downarrow} + F_{\uparrow}(W_{\downarrow} - V_{\downarrow}U_{\uparrow})]},$$

$$t_{\downarrow\uparrow} = \frac{1}{1 - F_{\uparrow}U_{\uparrow} - F_{\downarrow}[V_{\downarrow} + F_{\uparrow}(W_{\downarrow} - V_{\downarrow}U_{\uparrow})]} (-IS_+) \quad (2.22)$$

$$= (-IS_+) \frac{1}{1 - F_{\downarrow}U_{\downarrow} - F_{\uparrow}[V_{\uparrow} + F_{\downarrow}(W_{\uparrow} - V_{\uparrow}U_{\downarrow})]}.$$

Here, for the sake of simplicity, the t matrix elements in the Wannier representation $\langle m\mu | t | m\nu \rangle$ are written as $t_{\mu\nu}$, and $F_{\mu} \equiv F_{\mu}(\omega)$.

It should also be emphasized that this expression for the t matrix is very different from that derived by Nolting¹⁴⁾. This is because Nolting first calculated the thermal average for the fluctuating f spin to determine the quasi-particle energy levels, without considering the application of the CPA with an alloy analogy. Since the CPA should be used to treat the f spin fluctuation, Nolting's treatment is not suitable.

2.3 CPA conditions

The conditions for the CPA are expressed as

$$\langle t_{\uparrow\uparrow} \rangle_{\text{av}} = 0, \quad (2.23)$$

$$\langle t_{\downarrow\downarrow} \rangle_{\text{av}} = 0. \quad (2.24)$$

Since the matrix elements $t_{\uparrow\uparrow}$ and $t_{\downarrow\downarrow}$ also involve S_z only, the thermal average over the f spin states is easily calculated within the mean field theory (see also Appendix B in Ref. 5).

3. Results and Discussion

3.1 Overall features of the density of states

Since the present treatment is equivalent to Kubo's approximation⁶⁾, we add only a few comments concerning the self-energy (see also Appendix A in Ref. 5).

(a) Weak interaction limit: Inserting Eq. (2.19) into Eq. (2.23), and Eq. (2.20) into Eq. (2.24), and expanding them in I to the first order, we obtain

$$\langle V_{\uparrow} \rangle_{\text{av}} = 0 \text{ then } \Sigma_{\uparrow} = -I \langle S_z \rangle_{\text{av}}, \quad (3.1)$$

$$\langle V_{\downarrow} \rangle_{\text{av}} = 0 \text{ then } \Sigma_{\downarrow} = +I \langle S_z \rangle_{\text{av}}. \quad (3.2)$$

This result is equivalent to the one obtained using the first-order perturbation theory¹⁵⁾. Therefore, in the weak exchange interaction limit the present theory reproduces the simple spin-splitting band model, in which the conduction band splits into two spin-polarized subbands.

(b) Completely ferromagnetic case: In this case, only the value $S_z = S$ is realized in the thermal average over the f spin states. Hence we obtain, from Eqs. (2.23) and (2.24),

$$\Sigma_{\uparrow} = -IS, \quad (3.3)$$

and

$$\Sigma_{\downarrow} = IS \frac{(1 + IF_{\downarrow})}{(1 - IF_{\downarrow})}. \quad (3.4)$$

The results can be interpreted as follows. For the completely ferromagnetic case (i.e., $T=0$), the states of electrons with up-spin only shift $-IS$ with no damping. On the other hand, the s electron states with down-spin are damped because they can flip their spin under the condition that the total spin ($=S-1/2$) is conserved if the density of states with up-spin is not zero therein. This is because Σ_{\downarrow} involves $F_{\uparrow} (\equiv F_{\uparrow}(\omega))$.

The spin-flip process of the s electron with down-spin is a quantum effect due to the finiteness of the f spin value. Thus, in the classical spin limit (i.e., setting $S \gg 1$ and $S_z \gg 1$ in Eqs. (2.13)-(2.18)), Eq. (3.4) is replaced by

$$\Sigma_{\downarrow} = IS. \quad (3.5)$$

It is also verified that the self-energy given by Eqs. (3.3) and (3.4) is in accord with that obtained by Shastry and Mattis¹⁶⁾ for a single electron in a ferromagnetic semiconductor at $T=0$ if the magnon energy is negligible.

(c) Paramagnetic case: The expression for the paramagnetic temperature range (setting $\langle S_z \rangle_{\text{av}} = 0$, $\Sigma = \Sigma_{\uparrow} = \Sigma_{\downarrow}$ and $F = F_{\uparrow} = F_{\downarrow}$ etc.) agrees with that derived by Rangette *et al.*¹⁷⁾

In particular, for the strong exchange interaction limit (atomic limit), we obtain

$$\Sigma = \frac{I^2 S(S+1)}{\omega - I}, \quad (3.6)$$

and

$$F(\omega) \cong \frac{1}{\omega - \Sigma} = \frac{S+1}{2S+1} \frac{1}{\omega + IS} + \frac{S}{2S+1} \frac{1}{\omega - I(S+1)}. \quad (3.7)$$

This expression for $F(\omega)$ corresponds to the energy levels in the atomic limit being $-IS$ and $I(S+1)$, with degeneracies $2(S+1)$ and $2S$, respectively. This result is reasonable.

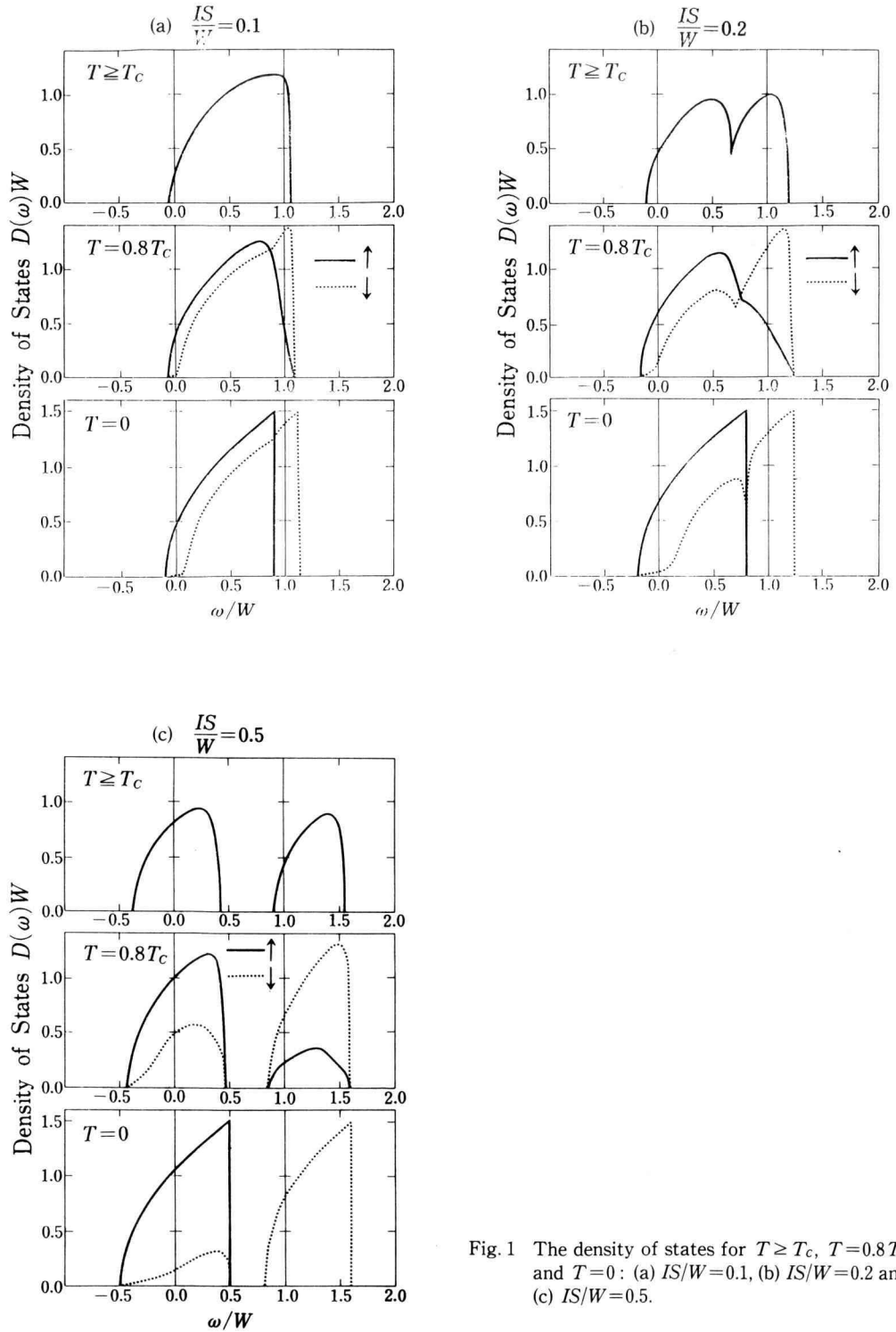


Fig. 1 The density of states for $T \geq T_c$, $T = 0.8T_c$ and $T = 0$: (a) $IS/W = 0.1$, (b) $IS/W = 0.2$ and (c) $IS/W = 0.5$.

As in Refs. 3-5, for the numerical calculation we simply assume a free-electron-like band with bandwidth W ; the energy of the Block band is

$$\varepsilon_k = W(k/q_D)^2 \quad \text{for } 0 \leq k \leq q_D, \quad (3.8)$$

where q_D is the radius of the Debye sphere. The summation over k in the first Brillouin zone in Eq. (2.12) is replaced by the integration within the Debye sphere. It is worth noting that introduction of a finite bandwidth (or $0 \leq \varepsilon_k \leq W$) is necessary to avoid the divergence in energy due to the point interaction (see Appendix C in Ref. 3). Furthermore, we set $S=7/2$ for Eu-chalcogenides. The thermal average for fluctuating f spin is calculated using the molecular field theory.

In Fig. 1 we show the density of states calculated in this study for three typical cases of the s - f exchange interaction strength: (a) weak interaction ($IS/W=0.1$), (b) intermediate interaction ($IS/W=0.2$) and (c) strong interaction ($IS/W=0.5$). Note that the values of the normalized magnetization $M \equiv \langle S_z \rangle_{\text{av}}/S$, calculated using the molecular field theory for $S=7/2$, are $M=0.0$ ($T \geq T_c$), $M=0.61$ ($T=0.8 T_c$) and $M=1.0$ ($T=0$).

We briefly point out a few characteristic features of the results. The first-order perturbation calculation shows that the ferromagnetic ordering of f spins gives rise to the $-I\langle S \rangle_{\text{av}}$ shift in the up-spin band and the $+I\langle S \rangle_{\text{av}}$ shift in the down-spin band. However, even for $IS/W=0.1$ this is not the case, as is shown in Fig. 1(a). The bands are broadened due to f spin fluctuation, and the down-spin band has a tail which reaches the bottom of the up-spin band even at $T=0$.

At high temperatures $T \geq T_c$, the band for $IS/W=0.2$ has a neck in the middle part (see Fig. 1(b)), and the band for $IS/W=0.5$ has two subbands (see Fig. 1(c)), which are characterized by the coupling of the electron spin parallel or antiparallel to the orientation of f spins. As the temperature decreases from T_c , the up-spin band is shifted to the low-energy side and diminishes at high energies where the states are mainly antiparallel-coupling states. In contrast, the down-spin band is shifted to the high-energy side on the whole, while the bottom of the down-spin band extends, accompanied by the lowering of the bottom of the up-spin band.

In all of the present numerical calculations, the total density of states is confirmed to be

$$\int_{-\infty}^{\infty} D_{\mu}(\omega) d\omega = 1.0 \quad (3.9)$$

for both $\mu = \uparrow$ and \downarrow .

3.2 Electron-spin polarization

Direct observation of spin-polarized conduction is obtained by the experiment of the electron-spin polarization. Kisker *et al.*¹⁸⁾ observed experimentally the spin polarization $P(T)$ of conduction electrons in EuS by means of field emission from a W-EuS junction, and revealed the strong temperature dependence of $P(T)$ (see the experimental results plotted in Fig. 2). According to the simple spin-splitting band model based on the first-order perturbation theory, the result should be $P(T)=1$ for $T < T_c$ and $P(T)=0$ for $T \geq T_c$. Thus, the observed spin polarization $P(T)$ of the field-emitted electrons is very different from that expected from the simple spin-splitting band model^{10,19)}.

In order to explain the experimental result, we assume that N_+/N_- is equal to $D_+(\omega)/D_-(\omega)$. Here $N_+(N_-)$ is the number of emitted electrons with up (down) spin,

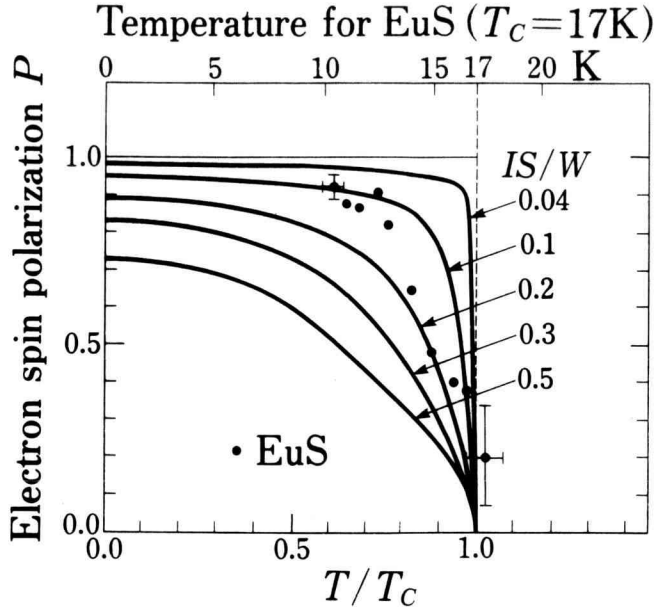


Fig. 2 The electron-spin polarization, P , is shown as a function of T/T_c for various values of IS/W . The experimental result for EuS reported by Kisker *et al* (Ref. 18) is also shown for comparison.

$D_+(\omega)(D_-(\omega))$ is the density of states for the conduction electrons with up (down) spin, and ω is a typical energy of the emitted electron. This assumption seems to be reasonable because the experiment was carried out under the condition that the conduction band was almost empty. Thus we have the following expression for the polarization, $P(T)^{20)}$.

$$P(T) = \frac{N_+ - N_-}{N_+ + N_-} = \frac{D_+(\omega) - D_-(\omega)}{D_+(\omega) + D_-(\omega)}. \quad (3.10)$$

In this study we set $\omega=0$ for simplicity.

In Fig. 2, the present result for $P(T)$ is shown as a function of the normalized temperature T/T_c for various values of IS/W , together with the experimental data for EuS¹⁸⁾. The result suggests that IS/W of EuS is between 0.1~0.2. Thus, the spin-filter experiment in EuS is explained satisfactorily within the framework of the singlet-site approximation.

4. Concluding Remarks

We aim to devise an improved theory for the s - f model, which is applicable for a wide range of IS/W and in a wide temperature range. However, it is too difficult to treat the scattering due to f spin correlation together with multiple scattering. Therefore, to take one step towards our goal, in this study, we have investigated the single-site approximation for the s - f model.

First, we derived the t matrix elements of the s - f exchange interaction for a single f spin embedded in the effective medium, where an s electron is subject to complex potential, Σ_+ or Σ_- , according to the orientation of its spin. Next, we studied the CPA conditions for the s - f model. The formula presented in this study is equivalent to Kubo's method⁶⁾, and is therefore in agreement with the CPA used by Rangette *et al.*¹⁷⁾ when the directions of f spins

are random, and it reproduces the result of the first-order perturbation theory¹⁵⁾ in the weak s - f interaction limit. Furthermore, our result agrees with that obtained by Shastry and Mattis¹⁶⁾ for a single electron in a ferromagnetic semiconductor at $T=0$, when the magnetic excitation energy is negligible. However, the present treatment is very different from that of Nolting and co-workers^{7-12,14)}, as shown in Sec. 2.

Assuming a free-electron-like band with bandwidth W for the s electron, numerical calculations were performed. The results for the density of states and the electron spin polarization were shown for various values of IS/W and for various temperatures. The temperature dependence of the electron-spin polarization, experimentally observed in ferromagnetic Eu-chalcogenides, were explained consistently within this theory.²¹⁾

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