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Single-site approximation for the s-f model in ferromagnetic semiconductors

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The conduction-electron state in a ferromagnetic semiconductor is studied theoretically by applying the coherent-potential approximation to the s-f model. In order to consider multiple scattering at ferromagnetic temperatures, we first derive the t-matrix elements of the s-f exchange interaction for a single f spin embedded in the effective medium, where a conduction electron is subjected to complex potential, $\Sigma_\uparrow$ or $\Sigma_\downarrow$, according to the orientation of its spin. 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. The anomalous redshift of the optical-absorption edge and the temperature dependence of the electron-spin polarization, experimentally observed in EuO and EuS, can be explained consistently within this approximation. [S0163-1829(96)00740-0]

I. 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.[1–3] 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.

$$H_t = H_s + H_f + H_{sf}, \tag{1.1}$$

$$H_s = \sum_{k\mu} \varepsilon_k a_{k\mu}^\dagger a_{k\mu}, \tag{1.2}$$

$$H_f = -\sum_{mn} J_{mn} \mathbf{S}_m \cdot \mathbf{S}_n, \tag{1.3}$$

$$H_{sf} = -I \sum_{m\mu\nu} a_{m\mu}^\dagger \boldsymbol{\sigma} \cdot \mathbf{S}_m a_{m\nu}. \tag{1.4}$$

The notations used here are the same as in the previous papers.[2,3]

A single electron (hereafter referred to as an 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,[2,3] we studied the variation of the density of states with temperature at paramagnetic temperatures, taking into account the scattering due to the 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 the 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 the f-spin correlation.

Pioneering work on the single-site approximation in the ferromagnetic temperature region was done by Kubo,[4] 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, a 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 co-workers.[5–9] Starting from the atomic limit solution, they first presented the quasiparticle multiband picture for the conduction band in magnetic semiconductors.[5–7] Using the quasiparticle levels and spectral weights calculated based on their concept, they formulated the CPA using an alloy analogy.[5,8,9] Their treatment, however, is somewhat questionable,[1] because the quasiparticle concept is fully realized only in the weak-coupling region, as they acknowledged in their study.[10] 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 the 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 organization of this paper is as follows. In Sec. II, 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. The actual procedure for the numerical calculation is given in Appendix A. It is proved in Appendix B that the present treatment is equivalent to Kubo's method. In Sec. III, the numerically calculated results for the density of states, the energy of the bottom of the conduction band, 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. IV, the concluding remarks are presented.

II. FORMALISM

A. 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] 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 the f spins.

Thus, we define the single-electron Green's function,[2,3]

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

with

$$H=H_s+H_{sf}, \tag{2.2}$$

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

In order to apply the multiple-scattering theory,[11] 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 an effective medium where an s electron is subject to complex potential, $\Sigma_\uparrow$ or $\Sigma_\downarrow$, according to the orientation of its spin. Then, an s electron moving in this effective medium is described by the (unperturbed) reference Hamiltonian K:

$$K=\sum_{k\mu}(\varepsilon_k+\Sigma_\mu)a^\dagger_{k\mu}a_{k\mu}. \tag{2.3}$$

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

$$V=\sum_m v_m, \tag{2.4}$$

with

$$v_m=\sum_{\mu\nu}a^\dagger_{m\mu}(-I\boldsymbol{\sigma}\cdot\mathbf{S}_m-\Sigma_\mu\delta_{\mu\nu})a_{m\nu}. \tag{2.5}$$

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

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

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

$$t_m=v_m[1-Pv_m]^{-1}. \tag{2.7}$$

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 theory,[11] the total scattering operator T, which is related to G as

$$G=P+PTP, \tag{2.8}$$

is expressed as the multiple-scattering series,

$$T=\sum_m t_m+\sum_m t_mP\sum_{n(\neq m)}t_n+\sum_m t_mP\sum_{n(\neq m)}t_nP\sum_{l(\neq n)}t_l+\cdots. \tag{2.9}$$

Within the single-site approximation, the condition

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

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

In this approximation, the density of states for the electron's spin μ (=↑ or ↓), $D_\mu(\omega)$, is calculated using

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

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

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

B. 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:

$$V_\uparrow=-IS_z-\Sigma_\uparrow, \tag{2.13}$$

$$V_\downarrow=+IS_z-\Sigma_\downarrow, \tag{2.14}$$

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

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

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

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

Here, S_z is the z component of the 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 ↑ (↓) 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 ↑ (↓) spin interacts with the 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 ↑ (↓) 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 straightforward and no more approximation is needed. The resulting expressions are

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

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

$$\begin{aligned}t_{\uparrow\downarrow}&=\frac{1}{1-F_\downarrow U_\downarrow-F_\uparrow[V_\uparrow+F_\downarrow(W_\uparrow-V_\uparrow U_\downarrow)]}\,(-IS_-)\\&=(-IS_-)\,\frac{1}{1-F_\uparrow U_\uparrow-F_\downarrow[V_\downarrow+F_\uparrow(W_\downarrow-V_\downarrow U_\uparrow)]},\end{aligned} \tag{2.21}$$

$$\begin{aligned}t_{\downarrow\uparrow}&=\frac{1}{1-F_\uparrow U_\uparrow-F_\downarrow[V_\downarrow+F_\uparrow(W_\downarrow-V_\downarrow U_\uparrow)]}\,(-IS_+)\\&=(-IS_+)\,\frac{1}{1-F_\downarrow U_\downarrow-F_\uparrow[V_\uparrow+F_\downarrow(W_\uparrow-V_\uparrow U_\downarrow)]}.\end{aligned} \tag{2.22}$$

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 Nolting.[12] This is because Nolting first calculated the thermal average for the fluctuating f spin to determine the quasiparticle 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.

C. CPA conditions

The conditions for the CPA are expressed as

$$\langle t_{\uparrow\uparrow}\rangle_{\rm av}=0, \tag{2.23}$$

$$\langle t_{\downarrow\downarrow}\rangle_{\rm av}=0. \tag{2.24}$$

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.

The actual procedure for numerical calculation is presented in Appendix A. The proof that the present approximation agrees with that obtained by Kubo[4] is shown in Appendix B. The present method shows good convergence and is applicable for incorporating the exchange scattering due to the f spin correlation.[3]

III. RESULTS AND DISCUSSION

A. Overall features of the density of states

Since the present treatment is equivalent to Kubo's approximation,[4] we add only a few comments concerning the self-energy.

(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

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

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

This result is equivalent to the one obtained using the first-order perturbation theory.[13] 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),

$$\Sigma_\uparrow=-IS, \tag{3.3}$$

and

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

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 $\Sigma_\downarrow$ involves $F_\uparrow$ [$\equiv F_\uparrow(\omega)$].

This 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 $S\gg1$ and $S_z\gg1$ in Eqs. (2.13)–(2.18)], Eq. (3.4) is replaced by

$$\Sigma_\downarrow=+IS. \tag{3.5}$$

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 Mattis[14] 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 temperatures (setting $\langle S_z\rangle_{\rm av}=0$, $\Sigma=\Sigma_\uparrow=\Sigma_\downarrow$, and $F=F_\uparrow=F_\downarrow$, etc.) agrees with that derived by Rangette, Yanase, and Kübler.[15]

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

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

and

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

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.

As in Refs. 2 and 3, for the numerical calculation we simply assume a single parabolic band with bandwidth W; the energy of the Bloch band is

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

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 an introduction of a finite bandwith (or $0 \leqslant \varepsilon_k \leqslant W$) is necessary to avoid the divergence in energy due to the point interaction (see Appendix C in Ref. 2). 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 $M \equiv \langle S_z \rangle_{\mathrm{av}}/S$, calculated using the molecular-field theory for $S=\frac{7}{2}$, are $M=0.0$ ($T \geqslant T_C$), $M=0.61$ ($T=0.8T_C$), 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 $-I\langle S \rangle_{\mathrm{av}}$ shift in the up-spin band and the $+I\langle S \rangle_{\mathrm{av}}$ 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 the 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 $T \geqslant T_C$, 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.[2,3] 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

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

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

B. Temperature dependence of the energy of the bottom of the conduction band

In Fig. 2, the energy of the bottom of the conduction band normalized by IS, ω_b/IS, calculated in this study, is shown as a function of T/T_C for various values of IS/W. For $T \geqslant T_C$, the energy of the bottom of the conduction band, ω_b, is obtained using[2]

$$\frac{\omega_b}{IS} = -\frac{6\left(\frac{IS}{W}\right)\left(1+\frac{1}{S}\right)}{1+3\left(\frac{IS}{W}\right)\left(\frac{1}{S}\right)+\sqrt{1+6\left(\frac{IS}{W}\right)\left(\frac{1}{S}\right)+36\left(\frac{IS}{W}\right)^2\left(1+\frac{1}{2S}\right)^2}}. \tag{3.10}$$

As the temperature decreases, accompanied by the development of spontaneous magnetization, ω_b decreases and reaches $-IS$ at $T=0$. The value of $-M$ ($\equiv -\langle S_z \rangle_{\mathrm{av}}/S$), shown by the dotted line, corresponds to the result of the first-order perturbation theory or the limit of the weak exchange interaction.

Using this figure we can easily explain the redshift of the absorption edge observed experimentally in ferromagnetic Eu chalcogenides. According to Wachter,[16,17] the total shift of the absorption edge amounts to about 0.26 eV (EuO), 0.18 eV (EuS), and 0.13 eV (EuSe). Since these optical-absorption bands are assigned to the $4f^7 \rightarrow 4f^6 5d(t_{2g})$ band transition, the redshift is ascribed to the lowering of the energy of the bottom of the d-like conduction band with the decrease in the temperature due to the d-f exchange interaction (see also later discussion).

The first-order perturbation theory gives $-I\langle S_z \rangle_{\mathrm{av}}$ as the energy of the bottom of the conduction band.[13] Although the first-order perturbation theory can explain the origin of the redshift, it cannot explain why these ferromagnetic semiconductors show such different redshifts despite the fact that they all have the same lattice structure, the same f spin value $S=7/2$, and almost the same exchange interaction energy IS.[6] In this study, ω_b is a function of IS/W at $T \geqslant T_C$ [see Eq. (3.10)], while $\omega_b = -IS$ (independent of W) at $T=0$. Since the exchange interaction has an intra-atomic character due to the strong localization of f electrons within the Eu^{2+} ion, the values of IS do not greatly differ in these Eu chalcogenides. Thus, the difference in the total redshift can be ascribed to the difference in the bandwidth W. For $IS=0.35$ eV, in this study, the observed redshift corresponds to the bandwidth $W=4.7$ eV (EuO), 1.9 eV (EuS), and 1.2 eV (EuSe). The agreement with the values for the $5d(t_{2g})$ conduction bandwidth obtained by Wachter[16] based on optical measurements seems reasonable.

Based on Fig. 2 we can also explain the reason for the apparent success of the first-order perturbation theory in explaining the magnetic redshift. The temperature dependence

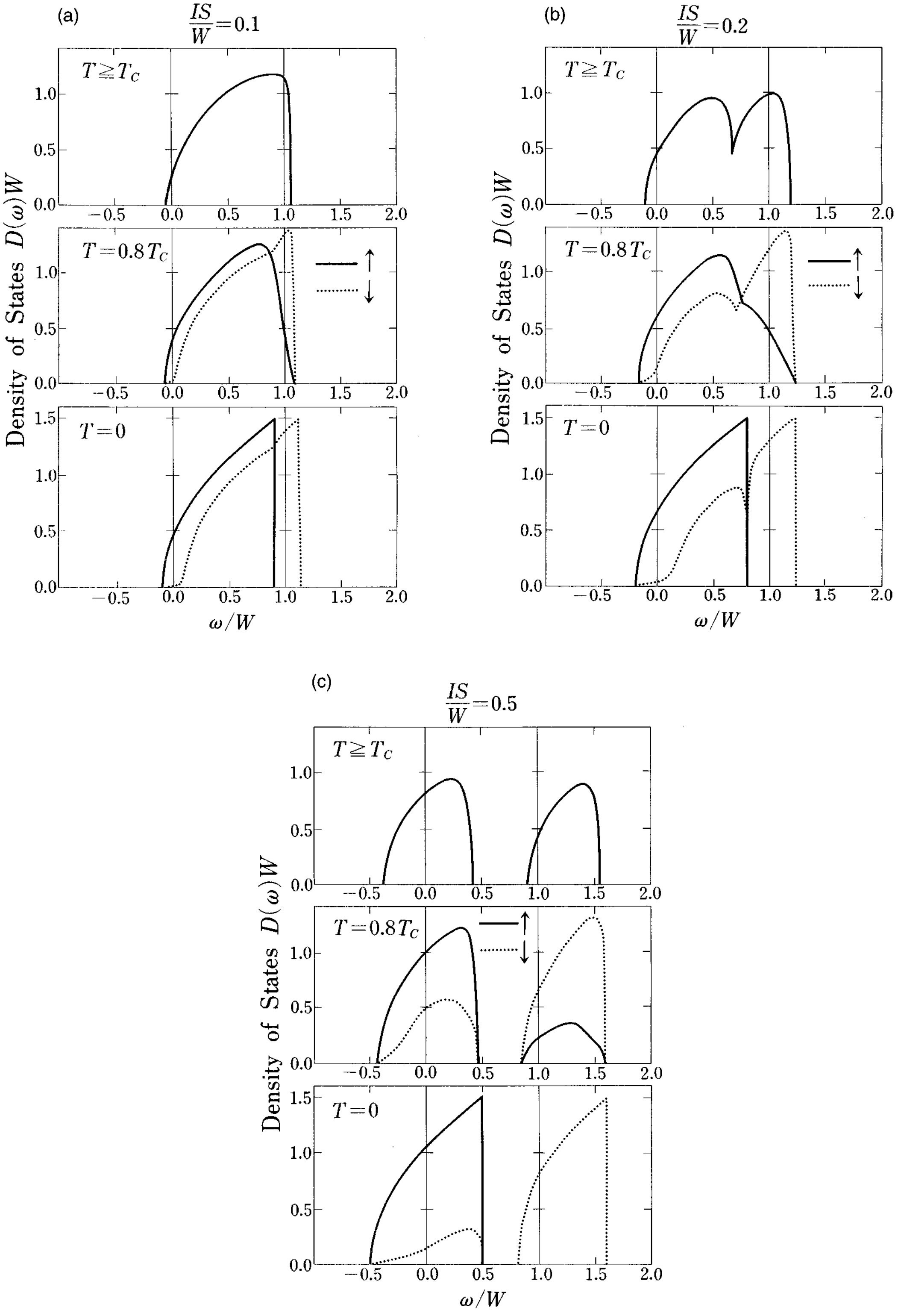

FIG. 1. The density of states for $T \geqslant T_C$, $T=0.8T_C$, and $T=0$: (a) $IS/W=0.1$, (b) $IS/W=0.2$, and (c) $IS/W=0.5$.

of the decrease in energy of the bottom of the band is described approximately as $-I_{\mathrm{eff}}\langle S_z\rangle_{\mathrm{av}}$, where I_{eff} is an effective exchange constant.

Upon incorporating the exchange scattering due to the f spin correlation, ω_b decreases further, especially at temperatures around T_C, but retains a finite value, in contrast with the result of the second-order perturbation theory,[18] as was previously reported by us.[3] Thus, incorporating the exchange scattering due to the f spin correlation will improve the agreement with the experimental results.[17]

In this study we attribute the optical-absorption band to the transition between the $4f$ level and the $5d$ conduction band.[1,16,17] However, there is another interpretation that the absorption spectra should be assigned to the $4f^7 \to 4f^6 5d(t_{2g})$ magnetic exciton.[19–22] In this interpretation an exciton consisting of a d electron and a hole in the f

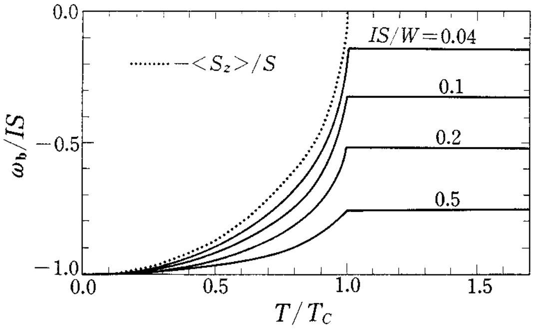

FIG. 2. The energy of the bottom of the conduction band normalized by IS, ω_b/IS, as a function of T/T_C for $IS/W=0.04$, 0.1, 0.2, and 0.5. The value of $-M$ $(\equiv -\langle S_z\rangle_{\mathrm{av}}/S)$, which corresponds to the result in the weak exchange interaction limit, is also shown as the dotted line for comparison.

level is formed after the transition. Even in that case, our conclusions are changed only slightly, as long as the excited electron spreads over many sites so that the energy of the electron behaves similarly to the bottom of the conduction band.[17]

C. Electron-spin polarization

Kisker *et al.*[23] 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. 3). 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 $T\geqslant T_C$. 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.[8,24]

In order to explain the experimental result, we assume that $N_\uparrow/N_\downarrow$ is equal to $D_\uparrow(\omega)/D_\downarrow(\omega)$. Here $N_\uparrow$ $(N_\downarrow)$ is the number of emitted electrons with up (down) spin, $D_\uparrow(\omega)$ $(D_\downarrow(\omega))$ 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, $P(T)$:[25]

$$P(T)=\frac{N_\uparrow-N_\downarrow}{N_\uparrow+N_\downarrow}=\frac{D_\uparrow(\omega)-D_\downarrow(\omega)}{D_\uparrow(\omega)+D_\downarrow(\omega)}. \tag{3.11}$$

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

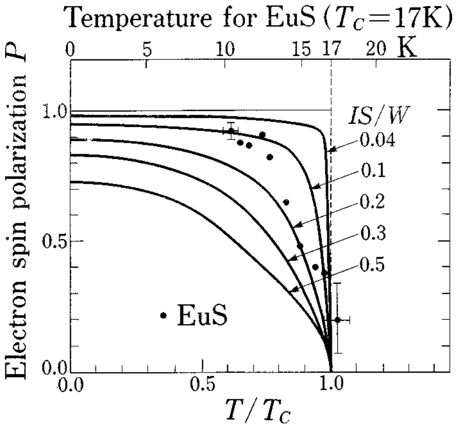

FIG. 3. The electron-spin polarization, P, is shown as a function of T/T_C for various values of IS/W. The experimental result for EuS taken from Kisker *et al.* (Ref. 23) is also shown for comparison.

In Fig. 3, 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 EuS.[20] The result suggests that IS/W of EuS is between 0.1–0.2, which is consistent with the result in the previous subsection. Thus, the spin-filter experiment in EuS is explained satisfactorily within the framework of the single-site approximation.

IV. 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 the 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, $\Sigma_\uparrow$ or $\Sigma_\downarrow$, 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 method,[4] and is therefore in agreement with the CPA used by Rangette, Yanase, and Kübler[15] when the directions of f spins are random, and it reproduces the result of the first-order perturbation theory[13] in the weak s-f interaction limit. Furthermore, our result agrees with that obtained by Shastry and Mattis[14] 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,[5–10,12] as shown in Sec. II.

Assuming a simple parabolic band with bandwidth W for the s electron, numerical calculations were performed. The results for the density of states, the energy of the bottom of the band and the electron-spin polarization were shown for various values of IS/W and for various temperatures. The anomalous redshift of the optical-absorption edge and the temperature dependence of the electron-spin polarization, experimentally observed in ferromagnetic Eu chalcogenides, were explained consistently within this theory.

The electronic resistivity for a ferromagnetic semiconductor was calculated based on the s-f model within the single-site approximation. The result seems reasonable, except at around T_C where exchange scattering due to the f spin correlation becomes significant, which will be reported by us in another paper.

An extension of the CPA to antiferromagnetic semiconductors was also attempted. The results of simple calculations indicated that an antiferromagnetic semiconductor shows a magnetic blueshift of the optical-absorption edge, which is consistent with the results of the experiment for EuTe.[16,17] Details will be reported elsewhere.

Throughout this study, we have ignored the effect of the f spin correlation. This effect plays an important role, especially at around T_C when IS/W is small, as for EuO. Incorporating the f spin correlation is our next subject of study.

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APPENDIX A

In the actual calculation for the CPA, the following method is adopted. For a given ω, assigning a suitable complex number to F_μ (for $\mu{=}\uparrow$ or $\downarrow$), we calculate the coefficients A_μ and B_μ for each spin orientation using the following definitions:

$$A_\uparrow = \left\langle \frac{S_z/S}{(1-F_\uparrow V_\uparrow)(1-F_\downarrow U_\downarrow)-F_\uparrow F_\downarrow W_\uparrow} \right\rangle_{\mathrm{av}}, \tag{A1}$$

$$B_\uparrow = \left\langle \frac{1}{(1-F_\uparrow V_\uparrow)(1-F_\downarrow U_\downarrow)-F_\uparrow F_\downarrow W_\uparrow} \right\rangle_{\mathrm{av}}, \tag{A2}$$

$$A_\downarrow = \left\langle \frac{S_z/S}{(1-F_\downarrow V_\downarrow)(1-F_\uparrow U_\uparrow)-F_\uparrow F_\downarrow W_\downarrow} \right\rangle_{\mathrm{av}}, \tag{A3}$$

$$B_\downarrow = \left\langle \frac{1}{(1-F_\downarrow V_\downarrow)(1-F_\uparrow U_\uparrow)-F_\uparrow F_\downarrow W_\downarrow} \right\rangle_{\mathrm{av}}, \tag{A4}$$

where $V_\uparrow$ ($V_\downarrow$), $U_\uparrow$ ($U_\downarrow$), and $W_\uparrow$ ($W_\downarrow$) are defined in Eqs. (2.13)–(2.18) in the text. Since these symbols include only S_z as an f spin operator, the thermal average for the fluctuating f spin can be easily calculated using the molecular-field theory at any temperature. Using A_μ and B_μ above, the CPA conditions, Eqs. (2.23) and (2.24), can be transformed into the equations to determine $\Sigma_\uparrow$ ($\Sigma_\downarrow$):

$$\Sigma_\uparrow = \frac{F_\downarrow I^2 S(S+1) - IS(1+F_\downarrow \Sigma_\downarrow)(A_\uparrow/B_\uparrow)}{1+F_\downarrow(\Sigma_\downarrow - I) - F_\downarrow IS(A_\uparrow/B_\uparrow)}, \tag{A5}$$

$$\Sigma_\downarrow = \frac{F_\uparrow I^2 S(S+1) + IS(1+F_\uparrow \Sigma_\uparrow)(A_\downarrow/B_\downarrow)}{1+F_\uparrow(\Sigma_\uparrow - I) + F_\uparrow IS(A_\downarrow/B_\downarrow)}. \tag{A6}$$

Once Σ_μ is determined, F_μ is obtained using Eq. (2.12) again. This procedure is repeated until the calculation converges. Our method converges far more quickly than does that of Kubo.[4]

APPENDIX B

Here we show that the present formula agrees with Kubo's CPA condition in terms of an effective locator, $\tilde{\mathcal{L}}_\sigma$. Setting $H_0 = F_0 = 0$ for simplicity, and replacing z by ω, $G^\sigma_{00}(z)$ by $F_\sigma(\omega)$, and $J^\sigma_0(z)$ by $\omega - \Sigma_\sigma - F_\sigma^{-1}$ in Eq. (15) of Ref. 4, we can obtain the following equation as the CPA condition:

$$F_\uparrow = \left\langle \frac{F_\downarrow^{-1} - U_\downarrow}{(F_\uparrow^{-1} - V_\uparrow)(F_\downarrow^{-1} - U_\downarrow) - W_\uparrow} \right\rangle_{\mathrm{av}}. \tag{B1}$$

Here F_σ, V_σ, U_σ, and W_σ ($\sigma{=}\uparrow$ or $\downarrow$) are defined by Eqs. (2.12)–(2.18) in the text. After a simple transformation, we can show that Eq. (B1) agrees with Eq. (2.23) into which Eq. (2.19) is substituted.

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[1] A. Mauger and C. Godart, Phys. Rep. **141**, 51 (1986).

[2] M. Takahashi, K. Mitsui, and M. Umehara, Phys. Rev. B **48**, 17 053 (1993); **50**, 17 759(E) (1994).

[3] M. Takahashi, K. Mitsui, and M. Umehara, Phys. Rev. B **52**, 16 313 (1995).

[4] K. Kubo, J. Phys. Soc. Jpn. **36**, 32 (1974).

[5] W. Nolting, Phys. Status Solidi B **96**, 11 (1979).

[6] W. Nolting and A. M. Oleś, Phys. Rev. B **22**, 6184 (1980).

[7] W. Nolting and M. Matlak, Phys. Status Solidi B **123**, 155 (1984).

[8] W. Nolting and A. M. Oleś, Solid State Commun. **33**, 961 (1980).

[9] W. Borgiel, W. Nolting, and G. Borster, Phys. Status Solidi B **136**, 131 (1986).

[10] W. Nolting, U. Dubil, and M. Matlak, J. Phys. C **18**, 3687 (1985).

[11] H. Ehrenreich and L. M. Schwartz, *Solid State Physics: Advances in Research and Application*, edited by H. Ehrenreich, F. Seitz, and D. Turnbull (Academic, New York, 1976), Vol. 31, p. 149.

[12] W. Nolting, Phys. Rev. B **32**, 403 (1985).

[13] C. Haas, in *New Developments in Semiconductors*, edited by P. R. Wallence, R. Harris, and M. J. Zuckermann (Noordhoff, Leyde, 1973), p. 1.

[14] B. S. Shastry and D. C. Mattis, Phys. Rev. B **24**, 5340 (1981).

[15] A. Rangette, A. Yanase, and J. Kübler, Solid State Commun. **12**, 171 (1973).

[16] P. Wachter, in *Handbook on the Physics and Chemistry of Rare Earths*, edited by K. A. Gschneidner, Jr. and L. Eyring (North-Holland, Amsterdam, 1979), p. 507.

[17] P. Wachter, CRC Crit. Rev. Solid State Sci. **3**, 189 (1972).

[18] C. Haas, Phys. Rev. **168**, 531 (1968).

[19] T. Kasuya and A. Yanase, Rev. Mod. Phys. **40**, 684 (1968).

[20] O. Sakai, A. Yanase, and T. Kasuya, J. Phys. Soc. Jpn. **42**, 596 (1977).

[21] M. Umehara, Phys. Rev. B **52**, 8140 (1995).

[22] R. Akimoto, M. Kobayashi, and T. Suzuki, J. Phys. Condens. Matter **8**, 105 (1996).

[23] E. Kisker, G. Baum, A. H. Mahan, W. Raith, and B. Reihl, Phys. Rev. B **18**, 2256 (1978).

[24] W. Nolting and B. Reihl, J. Magn. Magn. Mater. **10**, 1 (1979).

[25] M. I. Auslender and V. Yu. Irkhin, Solid State Commun. **50**, 1003 (1984).