Electron-spin polarization in ferromagnetic semiconductors

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Using the single-site approximation for the s-f model, the electron-spin polarization (ESP) in ferromagnetic semiconductors is studied theoretically. Assuming that the difference between the results of photoemitted ESP and those of field-emitted ESP is due to the difference in the energies of emitted electrons, the dependence of ESP on the temperature, external magnetic field, and energy, which is observed experimentally, can be explained consistently. [S0163-1829(97)01736-0]

I. INTRODUCTION

It is currently accepted that the condition band in ferromagnetic semiconductors, such as EuO and EuS, splits into two spin-polarized subbands below the Curie temperature (T_C).[1,2] Direct observation of the spin-split band is achieved by electron-spin polarization (ESP) measurement. Two methods have been used for the emission of electrons: photoemission (photo-ESP) and field emission (field-ESP). In the photo-ESP measurements,[3–8] the electrons absorbing light are emitted from the solid. On the other hand, in the field-ESP measurements, the W-EuS junction is used;[9–13] because the EuS conduction band is higher in energy than the W Fermi level, an external electric field is applied to tilt the bands such that the electrons from the W Fermi-level tunnel through the barrier into the empty EuS conduction band and finally reach the vacuum. The degree of polarization is measured using a Mott scattering analyzer, and expressed in terms of the ESP, P, defined by[3,4]

$$P=\frac{N_\uparrow - N_\downarrow}{N_\uparrow + N_\downarrow}, \tag{1.1}$$

where $N_\uparrow$ ($N_\downarrow$) is the number of emitted electrons with up (down) spin.

The main features of the measured ESP are summarized as follows.[1,2,4]

(i) On the whole, the degree of field-ESP observed is much higher than that of photo-ESP.

(ii) The applied magnetic-field dependence on ESP is very different between field-ESP and photo-ESP. Field-emitted electrons from EuS-coated W tips show spin polarization of up to 90% around T_C, even for a weak magnetic field such as 3 kOe. The observed field-ESP remains almost constant for a stronger magnetic field.[9] On the contrary, in the photo-ESP of EuS,[5] EuO,[6] La-doped EuO,[6,7] and Gd-doped EuO,[7] low values of ESP are observed even when the temperature is far below T_C, and the photo-ESP increases gradually with increasing magnetic field, but does not saturate at the expected strength of the magnetic field. Moreover, the photo-ESP of Gd-doped EuO, which is 4.3% at $T=43$ K, also remains low upon application of a magnetic field.[8]

(iii) Based on the variation of field-ESP as a function of the temperature T, the field-ESP appears to follow the normalized mean magnetization of the bulk in the range $0<T<T_C$, and reaches about 90% (not 100%) as $T\to 0$.[10,11] In contrast, the photo-ESP of Gd-doped EuO is considerably lower than the normalized bulk magnetization at low temperatures, and decreases monotonously as the temperature increases.[8]

(iv) In the field-ESP of EuS, the low-energy electrons are highly polarized, whereas the high-energy electrons are polarized only to a very low degree.[11–13] On the other hand, the photo-ESP of EuO, as a function of the incident photon energy, is about 20% for the photon energy of 3 eV, increases gradually to 30–40 % with an increase in photon energy, and then decreases for energies higher than 5 eV.[6]

Some theories have been proposed in order to account for these features in ESP. Since 100% spin-polarized $4f$ levels are well separated in energy from other bands, and in addition, are the highest occupied valence states, the electrons emitted from the $4f$ level are expected to be highly spin polarized at ferromagnetic temperatures or in a magnetic field. Nevertheless, the results of photo-ESP measurements are very different from those expected. Sattler and Sigemann[6] ascribed the low degree of photo-ESP to the depolarization that takes place at the surface layer before photoemitted electrons escape into the vacuum. They assumed the surface layer to be a paramagnetic sheet (dead layer). However, it is difficult to visualize why such a paramagnetic sheet is not active in the field-emission process.[2] Thus, the paramagnetic surface sheet cannot account for the high polarization observed in field-ESP. Furthermore, the experimental result shows that the spin-polarized band structure is the main factor that determines the dependence of photo-ESP on incident photon energy, and this also suggests the conservation of ESP in photoemission.[3] The reason for the low polarization in photo-ESP is more intrinsic, as will be shown herein.

Nolting and co-workers[14–17] discussed the temperature dependence of field-ESP on the basis of the quasiparticle multiband picture that was deduced from the atomic limit solution. They ascribed the behavior of ESP to the variations of the quasiparticle levels and the spectral weights calculated based on their concept. Their treatment is, however, somewhat questionable,[1,18] because the quasiparticle concept is fully realized only in the weak-coupling region.[19]

Considering the electron-magnon interaction, Edwards[18] presented a simple representation of the ESP as

$$P = \frac{\langle S_z \rangle_{av}}{S}, \tag{1.2}$$

which apparently agrees with experimental results in the field-ESP measurement.[1] However, Edwards's expression cannot account for the experimental results of photo-ESP.

Up to now, there has been no theory which consistently accounts for the observations of both photo-ESP and field-ESP. In this work, we calculate the electron-spin polarization based on the single-site approximation for the s-f model.[20] The difference between photo-ESP and field-ESP is explained by the difference in the energy of the emitted electrons.

The organization of this paper is as follows. In Sec. II, we present the model Hamiltonian and the actual procedure of the numerical calculation for the ESP. Furthermore, we present an explanation for the difference between field-ESP and photo-ESP. In Sec. III, the numerically calculated results for the dependence of the ESP on the magnetic field, the temperature, and the energy, are compared with experimental results. In Sec. IV, the concluding remarks are presented.

II. BASIC CONSIDERATION

A. Single-site approximation for the s-f model

The s-f exchange model is currently accepted as a basis for studying the conduction-electron states in ordinary magnetic semiconductors.[1,2] In this mode, 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 exchange interaction between f spins, and the s-f exchange interaction between an s electron and f spins, respectively,

$$H_t = H_s + H_f + H_{sf}, \tag{2.1}$$

$$H_s = \sum_{k\mu} \varepsilon_k a^\dagger_{k\mu} a_{k\mu}, \tag{2.2}$$

$$H_f = -\sum_{mn} J_{mn} \mathbf{S}_m \cdot \mathbf{S}_n - g\mu_B H_z \sum_m S_{mz}, \tag{2.3}$$

$$H_{sf} = -I \sum_{m\mu\nu} a^\dagger_{m\mu} \boldsymbol{\sigma} \cdot \mathbf{S}_m a_{m\nu}. \tag{2.4}$$

The notations used here are the same as in previous papers,[20] except that the applied magnetic field on the f spins is taken into account in H_f. In Eq. (2.3), a magnetic field H_z is assumed to be applied on the f spins in the z direction; g is the g factor and μ_B is a Bohr magnetron. The Zeeman effect on an electron is usually negligible.

In previous work,[20] we presented the single-site approximation for the s-f model. We first derived the t-matrix element of the s-f exchange interaction for a single f spin embedded in the effective medium, where an s electron is subjected to a complex potential, $\Sigma_\uparrow$ or $\Sigma_\downarrow$, according to the orientation of its spin. Next, we studied the coherent potential approximation (CPA) conditions for the s-f model. Furthermore, assuming a model band for the s electron, the density of states was calculated numerically.

In this work, we study the electron-spin polarization based on the single-site approximation for the s-f model. Thus, the usual mean-field theory is applied for f spins, ignoring the f-spin correlation. The details of the calculation for the density of states will not be repeated here. We only note the effect of an applied magnetic field on f spins. According to the molecular-field theory on a ferromagnet, the Curie temperature T_C is given by

$$T_C = \frac{2zJS(S+1)}{3k_B}, \tag{2.5}$$

where J $(=J_{mn})$ is an exchange integral, and z is the number of nearest neighbors of a magnetic ion. The molecular field is expressed by $2zJ\langle S_z\rangle_{av}/g\mu_B$. Thus, it is convenient to express the strength of the applied magnetic field using the normalized magnetic field h, which is defined by

$$h = \frac{g\mu_B H_z}{2zJS} = \frac{(S+1)g\mu_B H_z}{3k_B T_C}. \tag{2.6}$$

Note that $h=0.1$ corresponds to the magnetic field of 34.7 kOe for EuO ($T_C = 70$ K and $S=\frac{7}{2}$), and that of 8.43 kOe for EuS ($T_C=17$ K and $S=\frac{7}{2}$).

As in the previous study,[20] for the numerical calculation the energy of the undisturbed (model) band is assumed to be $\varepsilon_k = W(k/q_D)^2$ for $0 \leqslant k \leqslant q_D$, where q_D is the radius of the Debye sphere; the summation over k is replaced by the integration within the Debye sphere. In all of the present numerical calculations, the known number of states is confirmed to be

$$\int_{-\infty}^{\infty} D_\mu(\omega)\, d\omega = 1.0, \tag{2.7}$$

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

B. Consideration for the electron-spin polarization

We consider the relation between the density of states and the ESP.[3,4] When calculating the ESP, P, it is reasonable to assume that $N_\uparrow/N_\downarrow$ is equal to $D_\uparrow(\omega)/D_\downarrow(\omega)$, because the experiment was carried out under the condition that the conduction band was almost empty. Here, $D_\uparrow(\omega)$ $[D_\downarrow(\omega)]$ is the density of states for the energy ω of the emitted electrons with up (down) spin. Thus, we have the following expression for the ESP, P:[3,4]

$$P = \frac{D_\uparrow(\omega) - D_\downarrow(\omega)}{D_\uparrow(\omega) + D_\downarrow(\omega)}. \tag{2.8}$$

The numerical result is shown for $IS/W = 0.1$. Figure 1 illustrates the relation between the electric density of states, $D_\uparrow(\omega)$ and $D_\downarrow(\omega)$, and the ESP P, for $T=0.5T_C$. Note that $IS/W=0.1$ corresponds to the case of a weak s-f exchange interaction, and is appropriate for Eu chalcogenides (see also later discussion). The first-order perturbation for the s-f exchange interaction predicts that the ferromagnetic ordering of f spin gives rise to $-I\langle S_z\rangle_{av}$ shift in the up-spin band, and $+I\langle S_z\rangle_{av}$ shift in the down-spin band.[21] Thus, according to the simple spin-split band model based on the first-order perturbation theory, the result should be $P=100\%$ for $T<T_C$, and $P=0\%$ for $T \geqslant T_C$. However, even for $IS/W=0.1$ this is not the case, as shown in Fig. 1. The down-spin band has a tail which reaches the bottom of the up-spin band even at

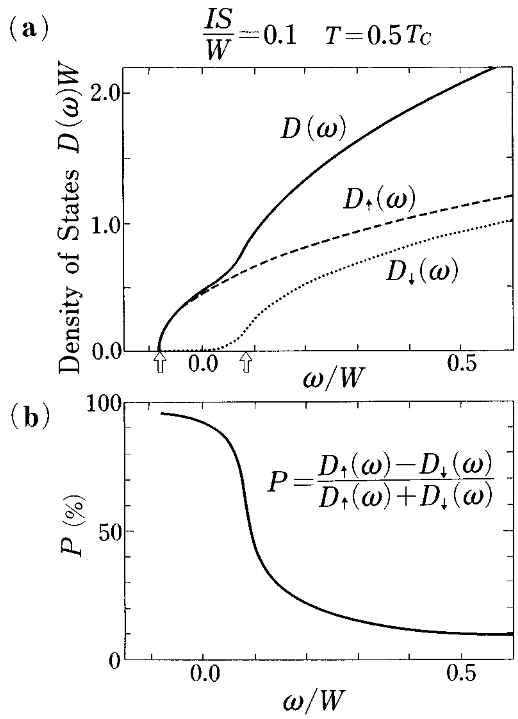

FIG. 1. (a) The products of the density of states and bandwidth, $D_{\uparrow}(\omega)W$, $D_{\downarrow}(\omega)W$, and $D(\omega)W$ $(=[D_{\uparrow}(\omega)+D_{\downarrow}(\omega)]W)$, and (b) the ESP, P, as a function of ω/W for $IS/W=0.1$ and $T=0.5T_C$. The open arrows (⇑) indicate the energies $\omega=-I\langle S\rangle_{av}$ and $\omega=+I\langle S\rangle_{av}$.

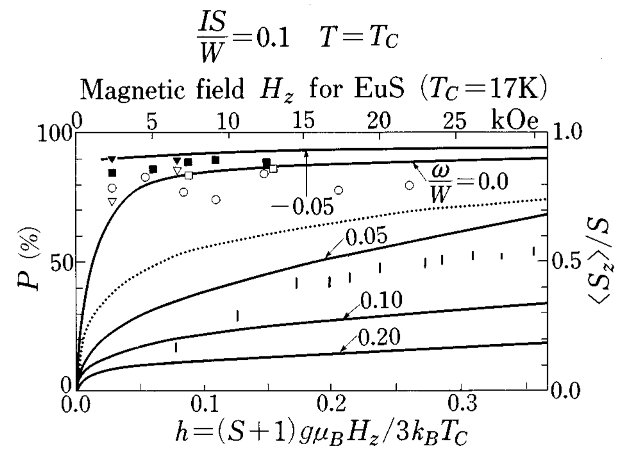

FIG. 2. The ESP at $T=T_C$, P, as a function of normalized applied magnetic field h, for $\omega/W=-0.05$, 0.0, 0.05, 0.10, and 0.20. The experimental results for the field-ESP of EuS (▽, □, ○ for 21 K, and ▼, ■ for 14 K) are taken from Müller *et al.* (Ref. 9), and the experimental results for photo-ESP on EuS (thick solid line) are taken from Bush, Campagna, and Siegmann (Ref. 5). The dotted line shows $\langle S_z\rangle_{av}/S$ calculated for $S=\frac{7}{2}$ using a molecular-field approximation.

$T=0$, which is a quantum effect due to the finiteness of the f-spin value.[20,22] Roughly speaking, for $-I\langle S_z\rangle_{av}<\omega<+I\langle S_z\rangle_{av}$, the electron spin is highly polarized, but does not reach 100% even at $T=0$. Around $\omega=+I\langle S_z\rangle_{av}$, the ESP decreases suddenly because the density of states with down-spin increases rapidly for $\omega>+I\langle S_z\rangle_{av}$. For still higher energy, the ESP decreases gradually.

Since EuS is an insulator, in the field-ESP measurement using the W-EuS junction, the EuS conduction bands are tilted when an external electric field is applied. If the field strength is sufficiently large, the bottom of the EuS conduction band is lowered to below the W Fermi level, and electrons can tunnel from W into the EuS conduction band and from there into vacuum. Therefore, the field-ESP corresponds to the values of P for energy equivalent to almost the bottom of the conduction band, such as $\omega/W=0$ in this case.

On the other hand, in the photo-ESP experiments, the energy distribution of light is usually chosen such that emission of electrons from $4f^7$ states is predominant.[3–8] The $4f$ electron absorbing a photon transfers to the conduction band; thus, most photoemitted electrons have energy related to a large density of states. Consequently, the electrons emitted during photo-ESP measurement have somewhat higher energy than the energy of the bottom of the band, such as $\omega/W=0.05-0.20$ in this case. As is shown through the present study, this interpretation for the ESP accounts consistently for the difference between photo-ESP and field-ESP.

III. RESULTS AND DISCUSSION

A. External magnetic-field dependence

In Fig. 2 we show the present result for the ESP at $T=T_C$, P, as a function of applied magnetic field for various values of the electron energy ω. For energies as low as $\omega/W<0.0$, the ESP is as high as 70–90 % even when the magnetic field is rather weak, and remains almost constant for still stronger magnetic fields. This feature explains well the measured field-ESP of EuS.[9] On the other hand, the ESP for somewhat higher energy, such as $\omega/W=0.05-0.20$, increases gradually with increasing applied magnetic fields, as if it does not show magnetic saturation. This feature agrees well with the measured photo-ESP of EuS.[5]

In Fig. 3 we show the result for the ESP at $T=0.5T_C$. At this temperature the localized f spins are highly magnetized even when no magnetic field is applied (see $\langle S_z\rangle_{av}/S$ shown by the dotted line). The ESP P calculated in this study, is almost independent of the strength of the applied magnetic field, while it is strongly dependent on the electron energy ω. In Fig. 3, the experimental results for the photo-ESP in $T=10$ K EuO (Ref. 6) and $T=43$ K 4.3% Gd-doped EuO (Ref. 8) are also included for comparison. In our understanding, photo-ESP corresponds to P for $\omega/W=0.05-0.20$. The calculated result satisfactorily accounts for the observations on the photo-ESP, except when a low magnetic field is applied. When the temperature is far below T_C, as in this case, the f spins in a ferromagnetic semiconductor are highly polarized spontaneously even when no magnetic field is applied. Thus, the applied magnetic field acts only to remove the domain structure.[4] Note that magnetic field $H_z>8$ kOe is needed for the saturation of the bulk magnetization in EuO.[1,4,6] Then, the discrepancy between the calculated result and the measured photo-ESP in a weak magnetic field will be ascribed to the domain effect.

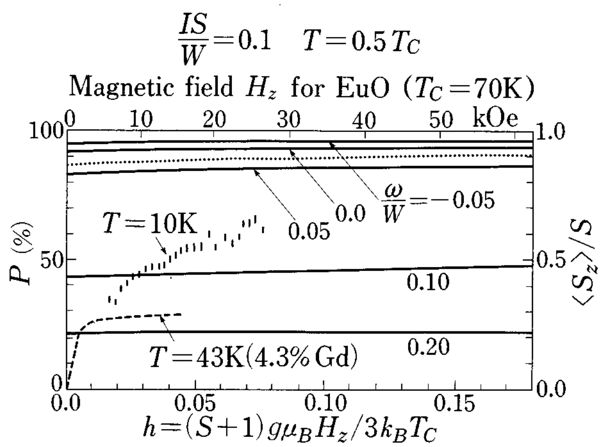

FIG. 3. The ESP at $T=0.5T_C$, P, as a function of h, for $\omega/W=-0.05$, 0.0, 0.05, 0.10, and 0.20. The experimental results for photo-ESP on EuO at 10 K (thick solid line) are taken from Siegmann (Ref. 4), and the experimental results for photo-ESP on 4.3 % Gd-doped EuO at 43 K are taken from Meier, Zürcher, and Kaldis (Ref. 8). The dotted line is $\langle S_z\rangle_{av}/S$ calculated for $S=\frac{7}{2}$.

B. Temperature dependence

In Fig. 4, we show the result for the ESP for $\omega/W=0.0$, P, as a function of normalized temperature T/T_C, for various values of IS/W, together with the field-ESP data for EuS.[11] Figure 4 also reveals the reasons for the apparent success of Edwards's expression, or Eq. (1.2). Furthermore, the result suggests that IS/W of EuS is between 0.1 and 0.2. Hereafter, we simply present the results for $IS/W=0.1$ (see also later discussion).

In Fig. 5 we show the ESP at $\omega/W=0.0$, P, as a function of T/T_C, for various strengths of the magnetic field applied. The field-ESP's of EuS in applied magnetic fields $H_z=0$, (Ref. 11) and 5 kOe (Ref. 10) are included for comparison. Note that normalized magnetic fields $h=0.05$ and 0.10 correspond to $H_z=4.2$ kOe and 8.4 kOe in EuS ($T_C=17$ K), respectively. The agreement between the calculated results and the measured field-ESP is satisfactory. When we use $IS/W=0.1$–0.2 instead of $IS/W=0.1$, the agreement with the observation will be further improved. These results strongly suggest that the field-ESP corresponds to the value of P for low electron energy, such as $\omega/W=0$ in this study, as already mentioned.

In Fig. 6 we show the ESP in the normalized magnetic field $h=0.03$, P, as a function of T/T_C for various values of electron energy ω/W. The photo-ESP's of 0.1% and 4.3% Gd-doped EuO are also included for comparison. Doping with Gd enhances the molecular field, and thus results in higher values of ESP. In this photo-ESP measurement, an external field of 10 kOe was applied in order to align the magnetic domains. Note that $h=0.03$ corresponds to 10 kOe for EuO ($T_C=70$ K). Comparison between the calculated results and observations suggests that photo-ESP corre-

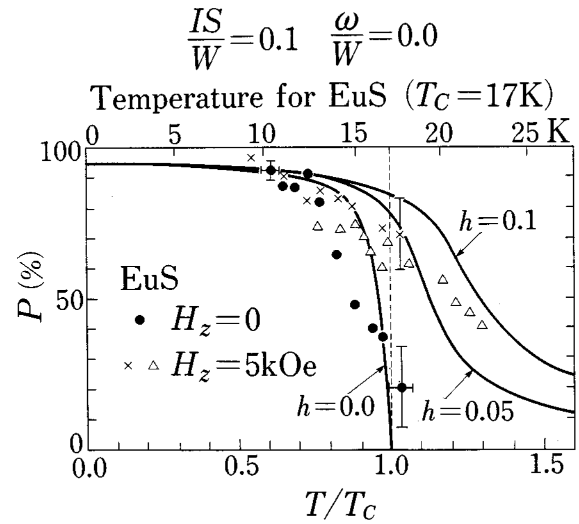

FIG. 5. The ESP, P, as a function of T/T_C for $h=0.0$, 0.05, and 0.10. Numerical results for P are obtained for $\omega/W=0.0$. The experimental results for field-ESP on EuS with no external magnetic field (●) and with a magnetic field of 5 kOe (×, △) are taken from Kisker *et al.* (Ref. 10).

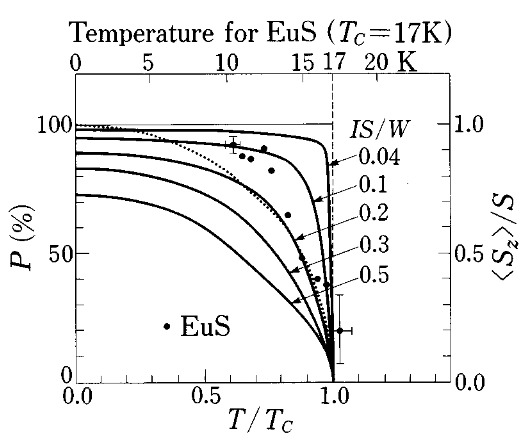

FIG. 4. The ESP for $\omega/W=0.0$, P, is shown as a function of T/T_C for various values of IS/W. The experimental results for field-ESP on EuS (●) are taken from Kisker *et al.* (Ref. 10), and the dotted line is for $\langle S_z\rangle_{av}/S$ calculated for $S=\frac{7}{2}$.

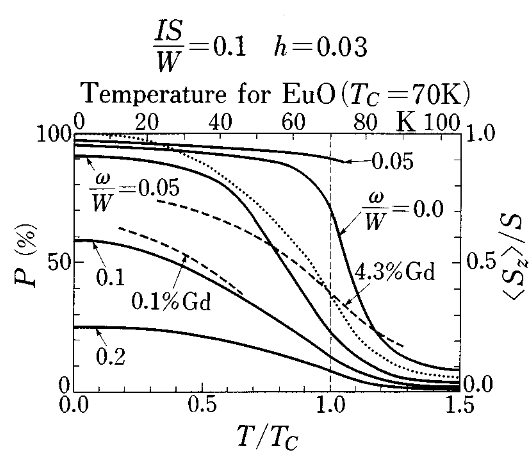

FIG. 6. The ESP, P, as a function of T/T_C for $\omega/W=-0.05$, 0.0, 0.05, 0.10, and 0.20. Numerical results are obtained for $h=0.03$. The experimental results for photo-ESP in an external field of 10 kOe on 0.1% Gd-doped EuO and 4.3% Gd-doped EuO (dashed line) are taken from Meier, Zürcher, and Kaldis (Ref. 8). The dotted line for $\langle S_z\rangle_{av}/S$ is also included for comparison.

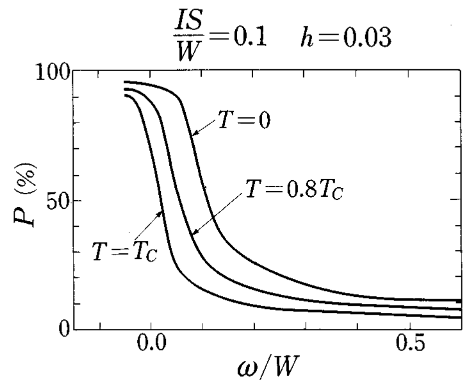

FIG. 7. The ESP, P, as a function of ω/W for $T=0$, $0.8T_C$ and $T=T_C$. Numerical results are obtained for $h=0.03$.

sponds to P for emitted electrons with energy somewhat higher than that of the bottom of the conduction band, such as $\omega/W=0.05-0.20$, in this study. This conclusion agrees with our view described above.

C. Energy dependence

In Fig. 7 we show the ESP in the applied magnetic field of $h=0.03$, P, as a function of the electron energy (ω), for various temperatures. The main features of the ω dependence of P are similar to those shown in Fig. 1, while the energy range in which the emitted electrons show high spin polarization is extended with the reduction of temperature. The field-ESP of EuS as a function of the filter-lens threshold energy showed that the low-energy electrons are highly polarized, whereas the degree of polarization of high-energy electrons is rather low.[10] Thus, the threshold energy dependence on the field-ESP is well explained using Fig. 7.

On the other hand, although the present result should be compared with the photo-ESP as a function of the energy of emitted electrons, no experiment that can be used as a reference has been reported, as far as we know. Thus, instead, the present result is compared with the photo-ESP as a function of the energy of incident photons.[6] The interpretation for the experimental result is as follows. For the photon energy $h\nu$ near the photothreshold (~2 eV), the photo-ESP shows low values, which is ascribed to uncontrolled impurity states; thus, the photo-ESP in this range of energies will disappear when samples having sufficiently good quality are measured. The photo-ESP becomes as high as 30–40 % for $h\nu \geqslant 4$ eV, indicating emission of electrons from the intrinsic $4f^7$ states, and decreases for $h\nu \geqslant 5$ eV. These experimental results are consistent with the present result for $\omega/W=0.05-0.6$, and strongly support our view described in the previous section. The absence of high ESP at low energies (near 4 eV) is because the current intensity of electrons photoemitted from $4f$ states is weak compared with that of electrons emitted from impurity states in the energy region. Again, we emphasize that in the photo-ESP experiments the energy distribution of light is usually chosen such that the dominant contribution to the photocurrent is from $4f$ electrons. For an instant, the photo-ESP of Gd-doped EuO (Ref. 8) was obtained at a photon energy of $h\nu=5$ eV, for which ω/W is roughly estimated to be 0.2.

IV. CONCLUDING REMARKS

Direct observations of the spin-polarized band were performed by electron-spin polarization (ESP) measurement. Nevertheless, there remain many riddles in the interpretation of the measured ESP. For example, the reason why the degree of field-ESP is much higher than that of photo-ESP has not been clarified. Furthermore, the differences in the dependencies of field-ESP and photo-ESP on the applied magnetic field, the temperature, and the energy have not been accounted for consistently.

In this study, based on the spin-polarized subband picture for ferromagnetic semiconductors that was previously proposed by us,[20] we attempted to explain the results of ESP measurements consistently. In Sec. II, we presented the procedure for calculating the ESP, P, together with the idea that measured field-ESP corresponds to P for the electron energy near the bottom of the conduction band, while measured photo-ESP corresponds to P for higher electron energy. Considering the process of electron emission, the interpretation seems reasonable. In Sec. III, in order to verify the spin-polarized subband picture and the newly proposed view for the ESP, we presented the numerical results for P as a function of the applied magnetic field, the temperature, and the electron energy, and compared them with the results of the ESP measurements. The agreement between the measured ESP and the present results is satisfactory.

In Fig. 4, the ESP for $\omega/W=0$, P in the present study, is shown as a function of the temperature, for various values of IS/W. On comparing the calculated result with the measured field-ESP of EuS,[11] we simply take $IS/W=0.1$ for Eu chalcogenides. Then, we present the various results obtained using $IS/W=0.1$, and compare them with the measured field-ESP and photo-ESP of the Eu chalcogenides. The results also suggest that $IS/W=0.1$ is appropriate for EuO, and $IS/W=0.1-0.2$ for EuS. This conclusion agrees with those obtained in our other work.[23]

In relation to Fig. 7, we note that the measurement of ESP is an effective method for investigating the band structure of magnetic semiconductors. The results are of value for comparison with the result obtained from spin-polarized band-structure calculation. Better experimental results will be obtained if, in addition to the ESP measurement, energy selection of the photoemitted electrons is performed for samples having a sufficiently good quality, which nowadays should be possible. Note also that in the ESP measurements EuO was a cleaved single crystal, while EuS was an evaporated polycrystalline film. Nevertheless, the present results consistently explain the ESP obtained experimentally. This suggests that the surface effect is not significant. However, the referred experiments were performed about 20 years ago, and samples might have involved uncontrolled impurities or imperfections. Thus, further experimental confirmation of the present theory is desired.

Throughout this work, we neglect the f-spin correlation which develops near T_C. So far, the singularity due to f-spin correlation has not been observed in the ESP. Thus, the f-spin correlation may not significantly influence the ESP.

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