Conduction electron states and ferromagnetism of electron-doped EuO

Masao Takahashi
Kanagawa Institute of Technology, Atsugi, 243-0292, Japan
E-mail: taka@gen.kanagawa-it.ac.jp

Abstract. To study the conduction electron states and ferromagnetism of electron-doped EuO theoretically, we have applied two approximations to the $s$-$f$ model and compared their results: virtual crystal approximation (VCA) and dynamical coherent potential approximation (dynamical CPA). The results of both approximations explain the anomalous magnetization curve experimentally observed in Gd-doped EuO and/or Eu-rich EuO with a low electron density, while only the result of dynamical CPA can explain the electron-density dependence of the Curie temperature $T_C$. The $T_C$ calculated by VCA shows the monotonous increase with electron density, while the $T_C$ calculated by dynamical CPA shows a maximum for a certain electron density. The mechanism of $T_C$ increase is also discussed.

Research interest in electron-doped EuO has been renewed in recent years with modern techniques and improved sample quality [1]. An increase in Curie temperature ($T_C$) and an unusual magnetization curve are observed not only in Gd-doped EuO [2] but also in La-doped EuO [3] and Eu-rich EuO [4]. Therefore, these phenomena are common in electron-doped EuO. In this study, basis of the $s$-$f$ model, we theoretically study the conduction electron states and ferromagnetism of electron-doped EuO. We apply virtual crystal approximation (VCA) and/or dynamical coherent potential approximation (dynamical CPA) to the $s$-$f$ model, and compare their results.

The Hamiltonian of the $s$-$f$ model is written as $H = H_s + H_f + H_{sf}$ [5, 6]. The first term $H_s$ represents the kinetic energy of a conduction ($s$-)electron. In this study, we employ the model density of states (DOS) of semicircular form with a half-bandwidth $\Delta = 3.5$ eV for the conduction $s$ band: $D_0(\omega) = \frac{2}{\pi\Delta} \sqrt{1 - \left(\frac{\omega}{\Delta}\right)^2}$. The second term $H_f$ represents the Heisenberg-type exchange interaction between $f$ spins of nearest neighbors. In the molecular field approximation (MFA), the Curie temperature $T_0$ is obtained as $k_B T_0 = 2zJS(S+1)/3$, where $J$ is the exchange integral between nearest $f$ spins, $z$ is the number of nearest-neighbor $f$ spins, and $S = 7/2$ for $f$ spins. We set $T_0 = 70$ K for pure EuO. The third term $H_{sf}$ represents the $s$-$f$ exchange interaction between an $s$ electron and $f$ spins: $H_{sf} = -I \sum_{m \mu \nu} a_\mu^\dagger \sigma_{\mu \nu} S_m a_{\nu}$. We set the $s$-$f$ exchange energy $IS = I \times S = 0.35$ eV (or $IS/\Delta = 0.1$).

We calculate the spin-polarized DOSs $D_\uparrow(\omega)$ and $D_\downarrow(\omega)$ for various values of $\langle S_z \rangle / S$, where $\langle S_z \rangle$ is the thermal average of the $f$ spin. Throughout the present work, we assume that the electron is degenerate. Then, the concentration of the electron with $\uparrow$ spin is calculated as $n_\uparrow = \int_{-\infty}^{\varepsilon_F} D_\uparrow(\omega) d\omega$. The Fermi level $\varepsilon_F$ is related to the electron concentration per site, $n$, by $n = n_\uparrow + n_\downarrow$. Thus, the electron energy per site is obtained as a function of $\langle S_z \rangle$ using
\[ E(\langle S_z \rangle) = E_\uparrow(\langle S_z \rangle) + E_\downarrow(\langle S_z \rangle) \]

with

\[ E_\uparrow(\langle S_z \rangle) = \int_{-\infty}^{\infty} \omega D_\uparrow(\omega) d\omega. \]  

(1)

The free energy per site of the system is given as a function of \( \langle S_z \rangle \) by \( F(\langle S_z \rangle) = E(\langle S_z \rangle) - \frac{1}{2}J(\langle S_z \rangle)^2 + TS \), where the entropy due to the localized spins is given by

\[ S = k_B \log \sum_{S_z=-S}^{S} \exp \left( \frac{hS_z}{k_B} - \frac{h}{T} \right) S_z. \]

First, by using the condition \( \frac{d}{d\langle S_z \rangle} F(\langle S_z \rangle) = 0 \), the effective magnetic field \( h \) is determined as

\[ h = h_1 + h_2 \]

where \( h_1 = 2zJ \langle S_z \rangle \) and \( h_2 = -\frac{d}{d\langle S_z \rangle} E(\langle S_z \rangle) \).

Next, we obtain \( \langle S_z \rangle \) as a function of temperature \( T \), by varying \( \langle S_z \rangle \) so as to give the minimum \( F(\langle S_z \rangle) \). We have verified that \( \langle S_z \rangle = SB_S(hS/k_BT) \) is satisfied, where \( B_S(x) \) is the Brillouin function. Furthermore, to gain insight into the origin and mechanism of electron-induced \( T_C \) increase, we also calculate the spin-coupling strength defined by

\[ Q(\omega) = \frac{\langle \delta(\omega - H) \sigma \cdot S \rangle / S}{\langle \delta(\omega - H) \rangle}. \]  

(3)

First, we present the results for the spin-polarized DOSs calculated with \( \langle S_z \rangle/S = 1.0, 0.5 \) and 1.0 by VCA and dynamical CPA in Figs. 1(a) and 2(a), respectively; the vertical lines indicate the positions of the Fermi level corresponding to the electron concentrations \( n = 0.01, 0.03, 0.05, 0.07, 0.10, 0.20 \) and 0.30. In VCA, the spin-polarized DOS is simply given by

**Figure 1.** Results obtained by VCA with \( \langle S_z \rangle/S = 1.0, 0.5 \), and 1.0. (a) Spin-polarized DOSs: \( D_\uparrow(\omega) \) and \( D_\downarrow(\omega) \). (b) Spin-coupling strength \( Q(\omega) \).

**Figure 2.** Results obtained by dynamical CPA with \( \langle S_z \rangle/S = 1.0, 0.5 \), and 1.0. (a) Spin-polarized DOSs: \( D_\uparrow(\omega) \) and \( D_\downarrow(\omega) \). (b) Spin-coupling strength \( Q(\omega) \).
When the magnetization changes from paramagnetic states (i.e., \(\langle S_z \rangle / S = 0\)) to completely ferromagnetic states (i.e., \(\langle S_z \rangle / S = 1\)), the Fermi levels shift to the lower-energy side, as is shown by the left arrows ("←") in the figures. The shift of the energy of the bottom of the band (called the "magnetic red shift") is \(IS = 0.35\) eV. The energy gain due to the development of ferromagnetism results in an increase in Curie temperature. The Curie temperature using the VCA is given by

\[
T_{C}^{\text{VCA}} = T_0 + \frac{2}{3} D_0 (\epsilon_F) \left( \frac{\Delta}{k_B} \right) \left( 1 + \frac{1}{S} \right) \left( \frac{IS}{\Delta} \right)^2.
\]  

\(T_{C}^{\text{VCA}}\) is a monotonically increasing function of \(n\) up to \(n = 1.0\), as shown in Fig. 3. In Fig. 1(b), we show the result for \(Q(\omega)\) calculated by VCA. In VCA, \(Q^{\text{VCA}}(\omega)\) corresponds to \(\langle \cos \theta \rangle\), where \(\theta\) is the angle between the spin of an \(s\) electron with an energy \(\omega\) and the localized \(f\) spin. When \(\langle S_z \rangle = 0\), \(Q^{\text{VCA}}(\omega) = 0\) because the directions of an \(s\) electron’s spin and \(f\) spins are assumed to be completely independent. In contrast, when \(\langle S_z \rangle = 1\), \(Q^{\text{VCA}}(\omega) = 1\) for \(-IS - \Delta \lesssim \omega \lesssim +IS - \Delta\) because all electron states are \(\uparrow\) spin states and the spins of the electron are coupled parallel to the \(f\) spins therein.

In dynamical CPA, the effect of the multiple-scattering of an \(s\) electron on an \(f\) spin due to \(s-f\) exchange interaction is taken into account. Thus, even when \(\langle S_z \rangle / S = 0\), the DOS does not agree with the model band \(D_0(\omega)\). The band is broaden and the bottom of the band of paramagnetic states is 0.08 eV lower than the bottom of the model band. As a consequence, the magnetic red shift is 0.27 eV (\(= 0.35 - 0.08\) eV). When \(n\) is small as \(n \lesssim 0.07\), the Fermi level decreases as the ferromagnetization develops, as shown in Fig. 2. When \(n\) is large as \(n \gtrsim 0.10\), however, the Fermi level shifts even to the higher-energy side. In Fig. 4, we show the Fermi levels \(\epsilon_F/\Delta\) calculated with various values of \(n\) as a function of \(\langle S_z \rangle / S\). Figure 4 shows that the energy gain due to the development of ferromagnetization increases when \(n \lesssim 0.05\), but decreases when \(n \gtrsim 0.10\). Thus, the \(n\) dependence of \(T_C\) obtained by dynamical CPA becomes like that shown in Fig. 3. With an increase in \(n\), \(T_C^{\text{D-CPA}}\) first increases, peaks at approximately \(n \approx 0.08\), and then decreases slowly. This suggests that there is an intrinsic limit to the electron-induced increase in the \(T_C\) of EuO [8]. Because the motion of the \(s\)-electron’s spin follows the motion of \(f\) spins to a
certain degree, $Q^{\text{D-CPA}}(\omega) \neq 0$ even when $\langle S_z \rangle = 0$, as shown in Fig. 2(b). When $\omega/\Delta \lesssim -0.83$, $Q^{\text{D-CPA}}(\omega)$ of $\langle S_z \rangle/S = 1$ is higher (lower) than that of $\langle S_z \rangle/S = 0$. Note that the change occurs at approximately the Fermi energy of $n$ of maximum $T_C$.

In Fig. 5, we show the result for magnetization $\langle S_z \rangle/S$ calculated by dynamical CPA as a function of $T$. In Fig. 6, we show the result for the normalized effective fields $h_2 S/\Delta$ as a function of $\langle S_z \rangle$. It is worth noting that in dynamical CPA, $h_2$ is not given by $h_2 = I(n_1 - n_\uparrow)$ but calculated using Eq. (2). The present result explains that the anomalous magnetization curve, which is generally observed in electron-doped EuO, is ascribed to the saturation of the effective field, as we have already discussed, applying VCA to the $s$-$f$ model [9].

To summarize, we have applied two approximations (VCA and dynamical CPA) to the $s$-$f$ model and compared their results. The results of both approximations explain anomalous magnetization curve experimental observation of electron-doped EuO with a low electron density $n$. The $T_C$ calculated by VCA is a monotonically increasing function of $n$. However, the $T_C$ calculated by dynamical CPA suggests the existence of an intrinsic limit to the increase in $T_C$ in electron-doped EuO. The decrease in $T_C$ with an increase in $n$ is explained by the fact that the spin of an $s$ electron with a high energy is exchange-coupled to $f$ spins stronger in paramagnetic states than in ferromagnetic states.

![Figure 5](image5.png) **Figure 5.** Magnetization $\langle S_z \rangle/S$ calculated with various values of $n$ by dynamical CPA as function of temperature $T$.

![Figure 6](image6.png) **Figure 6.** Normalized effective field $h_2 S/\Delta$ calculated with various values of $n$ by dynamical CPA as function of $\langle S_z \rangle/S$. The normalized effective field $h_1 S/\Delta = 2zJS\langle S_z \rangle/\Delta$ is included as $h(f\text{-spin})$.

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