Electrical resistivity in Gd-doped EuO and EuS

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Abstract

Based on the single-site approximation to s-f exchange model, the electrical resistivity in degenerate ferromagnetic semiconductors is studied theoretically. The spin-polarizeted conduction-band below the Curie temperature T_C is used for the determination of the Fermi energy. The result shows for a wide range of IS/W that the electrical resistivity due to s-f exchange interaction is constant when the temperature is higher than tha Curie temperature, and decreases to become zero at absolute temperature zero, where IS is the strength of an s-f exchange interaction and W is the width of a conduction band. The effect of external magnetic field is also investigated. The result explains the experimentaly observed magnetic resistance satisfactiorily.

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1. Introduction

The transport properties of magnetic semiconductors are unique in their strong dependence on the ordering of the localized magnetic moments[1,2]. The typical examples are Gd doped Eu chalcogenides. The carriers are supplied by dopants of Gd because the purest Eu chalcogenides are insulators. As a general rule, replacing a Eu^{2+} ion with a Gd^{3+} ion will result in a shallow donor. When the dopant and electron concentration is large the impurity band will be formed and it eventually merges with the bottom of the conduction band. This situation is realized when the doping exceeds 2% in EuO and SuS[3~6]. For these samples, the total resistivity $\rho_{tot}(T)$ is written as[3]

$$\rho_{tot}(T)=\rho_i+\rho_L(T)+\rho_m(T). \tag{1.1}$$

Here ρ_i is a temperature independent term due to neutral impurity scattering, $\rho_L(T)$ is due to lattice (acoustic phonon) scattering and is linear in temperature T, and $\rho_m(T)$ is a scattering term whose origin is the exchange interaction between the conduction electron and localized magnetic moments on each Eu ions (and/or Gd ions). An experimental curve for $\rho_m(T)$ is then obtained simply by subtracting ρ_i, the low temperature limit of the resistivity, and $\rho_L(T)$, which is determined from the high temperature slope, from the measured total resistivity, $\rho_{tot}(T)$. The remaining magnetic contribution, $\rho_m(T)$, is constant at high temperatures, takes maximaum near the Curie temperature T_C, and approaches zero with decreasing temperature.

At first appearance, the Born approximation (BA) for the resistivity in simple metals (Kasuya's theory[7]) seems to suffice for the explanation of the experimental results when the doping level is high enough to produce a degenerate semiconductor[3]. In this approximation, the exchange interaction is assumed to be weak and treated as a perturbation potential, or the conduction-band width is assumed to be broad. However, since the conduction bandwidth of EuO and EuS is controvertible[2,8], the application range for IS/W should be investigated in detil. Furthermore, in the Kasuya's theory, the Fermi energy ε_F is assumed to be much larger than the exchange energy IS : this is not the present case (see also later discussion).

On the other hand, the resistivity for strong exchange interaction was studied by K. Kubo

and N. Ohata[9] in connection with double exchange. When the exchange interaction is strong, the scattering mechanism is very different from the weak exchange interaction case. Since the electron has its spin pararell to the localized spin at each sites due to the strong exchange interaction, the hopping is strongly dependent on the relative orientation of the localized spins. Under the assumption that the exchange interaction is infinitetly strong, using the theory of Brownian motion and the virtual crystal approximation (VCA), they discussed the resistivity of $LaMnO_3$.

The reality may lie between these two limiting cases. However, as for as we kow, no unified theory for the resistivity has been presented, which is applicable to a wide range of IS/W. Therefore, it is desirable to build up an improved theory for the electron scattering due to the s-f exchange interaction. In the previous papers[10], we showed that the anomalous redshift of the opical absorption edge and the temperature dependence of the electron-spin polarization, experimentally observed in EuO and EuS, can be explained consistently within the single-site approximation. It this work, to extend the previous method, we adopt the Vericky theory[11] to the s-f model, to calculate the electrical resistivity of a degenerate ferromagnetic semiconductor.

2. Basic considerations

We treat the problem under the following simplications.

(a) We use the s-f exchange model. In this model there are magnetic moments on the regular sites and a well-defined conduction band. The conduction electron (s electron) moves in the crystal, while interacting with localized magnetic moments (f spins) through the s-f exchange interaction.

(b) A free-electron-like band with bandwidth W is assumed for the unperturbed s electron band.

(c) The f spin is treated by the molecular-field theory. Thus, the short-range ordering and the collective motion of f spins are ignored.

(d) The exisitence of the s electrons does not affect the f spins, while the s electron is scattered by the fluctuating f spins via s-f exchange interaction.

(e) The Coulomb interaction between s electrons is ignored.

In the above assumptions, (a)-(d) is the same as the previous study[10]. From the simplification (d) the electron localization due to exchange interaction (magnetic polaron problem) is out of our present scope. Thus, the concentration of conduction-electron is temperature independent, and the conductivity depends only on the mobility. When the s electron concentration is large, the Coulomb interaction between s electrons becomes important. Especially for the narrow band, the intra-atomic Coulomb repulsion due to the double occupation at the same site may become significant. By the assumption (e), however, this effect is ignored for simplicity. Thus, the s electrons are scattered at each lattice site independently. These situation correspond to doped single samples with doping levels of a few percent[1].

In the previous study[10], we studied the electron state in an effective medium, where an s electron is subjected to a complex potential, $\Sigma_\uparrow$ or $\Sigma_\downarrow$, according to the orientation of its spin ; and further, we calculated the density of tates, $D_\uparrow(\omega)$ and $D_\downarrow(\omega)$. When the concentration of s electrons, n, is given, thus, the Fermi energy ε_F is evaluated by using the spin-

polarized density of states. Then, the expression of the conductivity for electrons with spin μ is obtained straightforwardly by adopting the Vericky theory[11], as

$$\sigma_\mu=\frac{1}{A}\int_0^1 dx\left\{\mathrm{Im}\,\frac{Wx^2}{\varepsilon_F-Wx^2-\Sigma_\mu}\right\}^2. \tag{2.1}$$

Here, A is a constant which depends on the crystal structure, and defined by $A=3\pi^3 h/2e^2 q_D$ using the radius of the Debye sphere q_D.

The conductivity is easily evaluated when Σ_μ and ε_F are known. The electrical resistivity ρ is calculated by

$$\rho=\frac{1}{\sigma}=\frac{1}{\sigma_\uparrow+\sigma_\downarrow}. \tag{2.2}$$

3. Results and Discussion

The numerical calculation is performed for the s electron number per a site $x(=n/N)$ being 0.1. In Fig. 1, the resistivity for high-temperature limit, ρ_∞, is depicted as a function of IS/W. As shown in Appendix, when the exchange interaction is weak, the present result agrees with that of the Born approximation:[7,12~15]

$$\rho_\infty=3A\left(\frac{2}{x}\right)^{\frac{2}{3}}\left(\frac{IS}{W}\right)^2\left(1+\frac{1}{S}\right). \tag{3.1}$$

Thus, resistivity is proportional to $(IS/W)^2$ for small IS/W. The deviation from the Born approximation becomes large when $IS/W>0.1$[16]. The applicable range of the Born approximation is, thus, estimated as $IS/W<0.1$, which is consistent with that of the second-order perturbation[17,18]. For larger IS/W, the resistivity seems to approach a constant value.

Using Eq. (3.1) and the experimental data of Gd doped EuS[3], $n=2.2\times10^{20}\,\mathrm{cm}^{-3}$ (or $x=0.017$) and $\rho_\infty=5.6\times10^{-4}\,\Omega\cdot\mathrm{cm}$, together with $A=1.84\times10^{-5}\,\Omega\cdot\mathrm{m}$, we estimate as $IS/W=0.11$ (EuS). Besides, using the data of Gd doped EuO[5], $n=3.2\times10^{20}\,\mathrm{cm}^{-3}$ (or $x=0.011$) and $\rho_\infty=3.6\times10^{-4}\,\Omega\cdot\mathrm{cm}$, together with $A=1.58\times10^{-5}\,\Omega\cdot\mathrm{m}$, we estimate as $IS/W=0.09$ (EuO). This

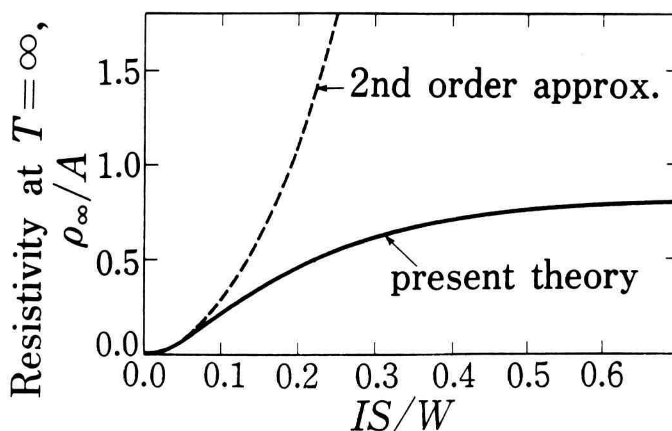

Fig. 1 The result for the (reduced) electrical resistivity at $T=\infty$, ρ_∞/A, as a function of the ratio of exchange strength to bandwidth, IS/W. The solid line for present theory and the dashed line for the Born approximation (BA).

suggests the value IS/W of EuS is larger than that of EuO, and probably suggests that the bandwidth W of EuO is larger than that of EuS because IS is common for all Eu chalcogenides. Further, Fig. 1 suggests that the EuO and SuS lie on the border of the applicable range of the Born approximation, and that the real values of IS/W may be a little larger than that obtained based on Eq.(3.1). These conclusions are consistent with previous interpretation for the anomalous redshift of the optical-absorption edge, and the temperature dependence of the electron-spin polarization, wherein IS/W of EuS is estimated to be 0.1 ~0.2[10].

In Figs. 2 and 3, (a) the ratio of the up-spin electron concentration, $n_\uparrow/(n_\uparrow+n_\downarrow)$, and (b) the resistivity ρ/A, are shown as a function of the normalized temperature T/T_C. We also studied the effect of the external magnetic field. In magnetic semiconductors, a magnetic field changes the f spin ordering. This results in the change of the electronic density of states, and influences the resistivity. The normal magnetoresistance, which occurs also in non-magnetic semiconductors, is usualy quite small and negligible. In this study, thus, we assume that the external magnetic field H_z affects only the f spins, or, the external fiend is taken into account when the thermal average over f spin states is performed. The results for various normalized magnetic fields, h, defined by

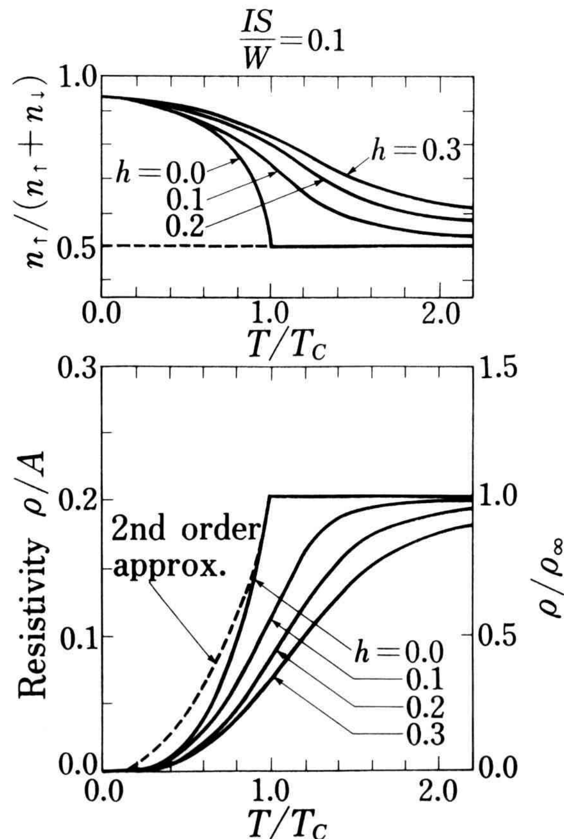

Fig. 2 The result for $IS/W=0.1$ under the normalized external magnetic field h as a function of normalized temperture T/Tc : (a) the ratio of $n_\uparrow$ to $n_\uparrow+n_\downarrow$ (b) the (reduced) electrical resistivity ρ/A. The result of the Born approximation (BA) for ρ/ρ_∞ is shown by the dashed line.

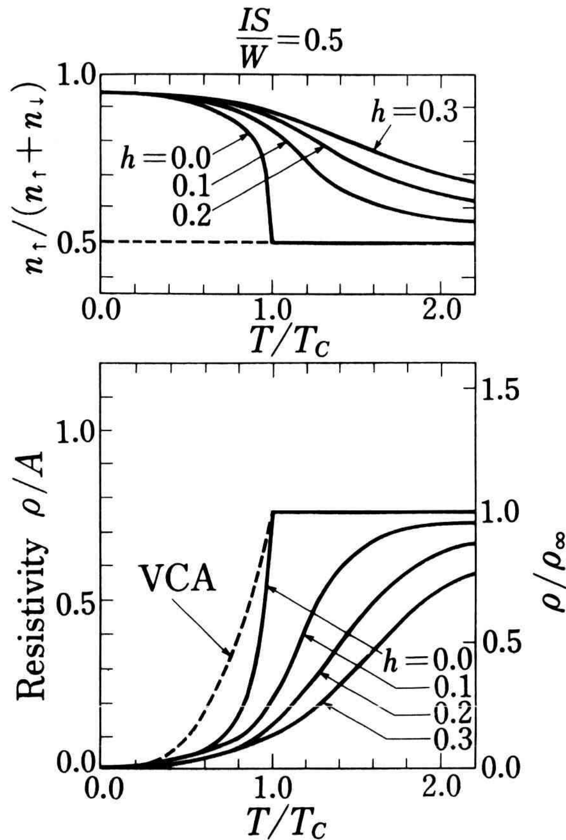

Fig. 3 The result for $IS/W=0.5$ under the external magnetic field h as a function of normalized temperture T/T_C : (a) the ratio of $n_\uparrow$ to $n_\uparrow+n_\downarrow$ (b) the (reduced) electric resistivity ρ/A. The result of the virtual crystal approximation (VCA) for ρ/ρ_∞ is shown by the dashed line.

$$h=\frac{(S+1)g\mu H_z}{3k_B T_C}, \tag{3.2}$$

are also shown in these figures.

Figure 2 is the result for $IS/W=0.1$, a typical case of a weak exchange interaction. For comparison, the result obtained using the Born apprroximation (BA) for resistivity when $h=0$[7],

$$\frac{\rho}{\rho_\infty}=\frac{1}{S(S+1)}\{S(S+1)-\langle S_z\rangle_{\rm av}^2-\langle S_z\rangle_{\rm av}\}, \tag{3.3}$$

is inserted. Note that Kasuya's theory is derived assuming that $n_\uparrow=n_\downarrow$ even for $T<T_C$. This is not true even for $x=0.1$ and $IS/W=0.1$, as shown in Fig. 2(a). We also shown the effect of external magnetic field. The obtained result seems reasonable[3].

The similar result for $IS/W=0.5$, as a typical case of a strong exchange interaction, is shown in Fig. 3. For comparison, the result derived on the basis of the virtual crystal approximation (VCA) for resistivity[9], is inserted.

4. Conculding Remarks

In real ferromagnetic semiconductors, a large peak around T_C in the resistivity-versus-temperature curve is observed, which is due to the electron scattering by the f spin correlation. For the case of weak exchange interaction, Haas[12] caluculated the mobility using the second-order perturbation, togther with the f spin correlation in terms of susceptibility $\chi(k)$ of $k \rightarrow 0$. The results were good except for a T_C, and in good agreement with Eq. (3.1) at high-temperature limit. However, for degenerate semiconductors, the k dependence of $\chi(k)$ cannot be neglected[13,14].

We aim to devise an improved theory based on the s-f model for the resistivity of a degenerate ferromagnetic semiconductor, which is applicable in a wide range of IS/W and in a wide temperature range. In the improved theory, the scattering due to f spin correlation should be treated simulteniously together with multiple scattering on one site. However, it is very difficult. Therefore, in this study to take one step towards the final goal, by extending the single-site approximation[10] we adopt the Vericky theory to s-f model, to calculate the resistivity of a degenerate ferromagnetic semiconductor. The present treatment is ineffective near T_C because the correlation of f spins is ignored throughout this work. However, the present method is effective for a wide range of IS/W and a wide temperature range except for near T_C.

Throughout this work, the carrier concentration (or the number of s electrons) is assumed to be temperature independent. This assumption may not suitable for the case of low doping as $x<0.02$[1], For such lightly doped samples, the electrical transport properties may be interpreted in terms of hopping between an empty impurity state and an occupied (magnetic) impurity state[3,19].

In real samples, as the quantity of doping increases, the ordering temperature rises up. This is due to the indirect exchange via free carriers[2]. Although this effect is not teated by the assumption (d) in this study, it may be partly incorporated by treating T_C as a parameter, or by introducing an effective field h in this work.

It should also be noted that at sufficiently low temperatures the method of the molecular field is not appropriate, and we should use the spin-wave method.

A Appendix A

Here we show that in the limit of weak exchange interaction the present formula at paramagnetic temperatures agrees with that of Born approximation. At paramagnetic temperatures, $\Sigma=\Sigma_\uparrow=\Sigma_\downarrow$, and in this case, $\left|\frac{\Sigma}{W}\right| \ll 1$. Thus from Eq. (2.2)

$$\sigma=2\sigma_\uparrow=2\sigma_\downarrow=\frac{ne^2\tau}{m}, \tag{A1}$$

with

$$\frac{\hbar}{\tau}=\frac{1}{2\,|\mathrm{Im}\Sigma|}. \tag{A2}$$

Putting the result of the second-order pertuebation (or of the limit of weak exchange interaction in the CPA),

$$\left|\frac{\mathrm{Im}\Sigma}{W}\right| = \frac{3}{2}\pi\left(\frac{\varepsilon_F}{W}\right)^{\frac{1}{2}}\left(\frac{IS}{W}\right)^2\left(1+\frac{1}{S}\right) \tag{A3}$$

into Eq. (A.2), we obtain Eq. (3.1) in the text.

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