

## Effects of $s$ - $d$ Interaction on Transport Phenomena

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The effects of the so-called  $s$ - $d$  interaction on transport phenomena of ferromagnetic metals and alloys are discussed. Electrical and thermal conductivities and thermoelectric effect are calculated with the simple molecular field approximation, and an anomalous thermoelectric effect is expected in these substances. In sufficiently low temperatures, the disordering of spin system is described by the collective mode of spin wave. The calculations of transport coefficient by using the spin wave approximation are also carried out.

### § 1. Introduction

In ferromagnetic metals and alloys the most characteristic interaction is the so-called  $s$ - $d$  interaction, namely, the spin exchange interaction between the conduction and the unfilled inner shell electrons (hereafter referred to as  $c$ - and  $u$ -electrons respectively). One of the main effects of this interaction is the effective spin exchange interaction between  $u$ -electrons which were treated firstly by Zener<sup>1)</sup> and in detail by us<sup>2)</sup> and Yosida<sup>3)</sup>. The other one of the main effects is the scattering mechanism of  $c$ -electrons which has the important contribution to the transport phenomena. The effects on the electrical resistivity are already treated by us<sup>4)</sup> for ferromagnetic metals and by Yosida<sup>5)</sup> for dilute alloys using the simple molecular field approximation. The problems concerning the effect on the superconductivity we treated in another paper<sup>6)</sup>. Here we treat the effects of  $s$ - $d$  interaction on the thermal resistivity and the thermoelectric force using the molecular field approximation. The anomalous thermoelectric force is expected to occur in these substances. The thermal conductivity by the spin diffusion seems to be also important because of the long range character of the  $s$ - $d$  interaction. Furthermore the transport coefficient in sufficiently low temperatures are also calculated with the use of the spin wave approximation.

### § 2. Fundamental standpoints

To solve the Boltzmann Equation rigorously, because for the discussion of the thermoelectric force careful treatment is required, we use here the following simplified model.

(i) We assume that  $c$ -electron is represented by one band model with effective mass  $m$  and that the effect of  $s$ - $d$  interaction is represented by the effective exchange

field  $H_c$ . Then the energy spectrum of  $c$ -electron is written as

$$E_k^\pm = \frac{\hbar^2 k^2}{2m} \mp H_c, \quad (1)$$

where  $H_c$  is approximated by

$$H_c = (N_i/N) (g-1) \bar{j}_z J(0), \quad (2)$$

where  $\bar{j}_z$  means the mean value of the  $z$ -component of the total angular momentum  $j_{nz}$  located in each magnetic ion,  $J(0)$  the  $s$ - $d$  exchange integral,  $N_i$  the number of magnetic ions per unit volume, therefore in metals  $N_i = N$ , and  $g$  the Lande's  $g$ -factor. In most cases,  $H_c$  is much smaller than the Fermi energy  $\zeta$ .

(ii) The other scattering mechanisms than the  $s$ - $d$  exchange interaction, such as electron-phonon and electron-non magnetic impurity atoms, are neglected in our treatment. These effects are discussed in later sections.

(iii) The Hamiltonian of the  $s$ - $d$  interaction is given by

$$\begin{aligned} H_{sd} = & (g-1)/N \cdot \sum_k \sum_{k'} \sum_{R_n} \exp\{i(\mathbf{k}-\mathbf{k}')\mathbf{R}_n\} [\{A(|\mathbf{k}-\mathbf{k}'|)/(g-1) - J(|\mathbf{k}-\mathbf{k}'|)j_n^z\} \\ & a_{k+}^* a_{k'+} + \{A(|\mathbf{k}-\mathbf{k}'|)/(g-1) + J(|\mathbf{k}-\mathbf{k}'|)j_n^z\} a_{k-}^* a_{k'-} \\ & - J(|\mathbf{k}-\mathbf{k}'|)j_n^- a_{k+}^* a_{k'-} - J(|\mathbf{k}-\mathbf{k}'|)j_n^+ a_{k-}^* a_{k'+}], \end{aligned} \quad (3)$$

where  $a_{k\nu}$  and  $a_{k\nu}^*$  mean respectively the creation and annihilation operators of electron with wave vector  $\mathbf{k}$  and spin quantum number  $\nu = \pm 1/2$ ,  $J(q)$  the exchange integral and  $A(q)$  the interaction except the exchange integral.

(iv) The energy level of  $\{j_n\}$  may be very complicated both in metals and dilute alloys. However, we use here the most simplified model that the energy level of  $j_n$  is represented by the single molecular field  $H_0$  such as

$$H_{\text{mag}} = - \sum_n H_0 j_n^z, \quad (4)$$

and thus the distribution of  $j_n^z$  is given by

$$w(j_z) = \exp(H_0 j_z / \kappa T) / \sum_{j_z} \exp(H_0 j_z / \kappa T). \quad (5)$$

(v) The scattering matrix element is given by the Born approximation. The transition probability, where the spin direction of  $c$ -electron does not change, namely, the first two processes of Eq. (3), is given by

$$W_\pm(q) = \begin{cases} N^{-1}(g-1)^2 |J(q)|^2 \{\bar{j}_z^2 - j_z^2\}, & \text{in metals} \\ N_i/N^2 \{|A(q)|^2 + (g-1)^2 |J(q)|^2 \bar{j}_z^2 \mp 2(g-1) \text{Re}(J(q)A(q))\bar{j}_z\}, & \text{in dilute alloys,} \end{cases} \quad (6)$$

where  $\bar{j}_z$  and  $\bar{j}_z^2$  mean the values of  $j_z$  and  $j_z^2$  averaged by using Eq. (5). Furthermore, we assumed that  $(\bar{j}_z - j_z^2)$  and in dilute alloys the positions of magnetic ions are perfectly at random and have no correlation in each other. On

the other hand, the transition probabilities, where the spin directions change each other, namely, the last two processes of Eq. (3), are given by

$$V_{\pm}(q) = N_i/N^2(g-1)^2 |J(q)|^2 \{j^2 + j - \bar{j}_z^2 \pm \bar{j}_z\}, \quad (7)$$

where  $\pm$  mean respectively the processes in which the spin direction of *c*-electron becomes  $\pm$ . Also in the above treatment we assumed that there are no correlation between magnetic ions. It is noticeable that between  $V_{\pm}(q)$  there is a relation

$$V_+(q) = e^x V_-(q), \quad (8)$$

where

$$x = H_0/\kappa T. \quad (9)$$

Using these assumptions, both the drift and collision terms of Bloch equations concerning *c*-electrons with  $\pm$  spin directions are written as

$$(\partial f_{\pm}(\mathbf{k})/\partial t)_{\text{drift}} = f_1(\varepsilon_{\pm}) v_x/\kappa T \cdot [(e\mathbf{F} - \nabla\zeta) - \varepsilon_{\pm} \kappa \nabla T], \quad (10)$$

$$\begin{aligned} (\partial f_{\pm}(\mathbf{k})/\partial t)_{\text{coll}} = & 2\pi/\hbar \cdot \sum_{\mathbf{k}'} [W_{\pm}(|\mathbf{k}-\mathbf{k}'|) \{f_{\pm}(\mathbf{k}') (1-f_{\pm}(\mathbf{k})) \\ & - f_{\pm}(\mathbf{k}) (1-f_{\pm}(\mathbf{k}'))\} \delta(E_k^{\pm} - E_{k'}^{\pm}) \\ & + \{V_{\pm}(|\mathbf{k}-\mathbf{k}'|) f_{\mp}(\mathbf{k}') (1-f_{\pm}(\mathbf{k})) \\ & - V_{\mp}(|\mathbf{k}-\mathbf{k}'|) f_{\pm}(\mathbf{k}) (1-f_{\mp}(\mathbf{k}'))\} \delta(E_k^{\pm} - E_{k'}^{\mp} \pm H_0)], \end{aligned} \quad (11)$$

where the directions of the electric field  $\mathbf{F}$  and the temperature gradient  $\nabla T$  are chosen in the *x* axis,  $f_1(\varepsilon)$  means

$$f_1(\varepsilon) = [(e^{\varepsilon} + 1)(1 + e^{-\varepsilon})]^{-1}, \quad (12)$$

and

$$\varepsilon_{\pm} = (E_k^{\pm} - \zeta)/\kappa T. \quad (13)$$

Further considering that  $f_{\pm}(\mathbf{k})$  is written as

$$f_{\pm}(\mathbf{k}) = f_0(\varepsilon_{\pm}) + k_x C_{\pm}(\varepsilon_{\pm}) f_1(\varepsilon_{\pm}), \quad (14)$$

where

$$f_0(\varepsilon) = [e^{\varepsilon} + 1]^{-1}, \quad (15)$$

and restricting to the linear theory, the collision terms become

$$\begin{aligned} (\partial f_{\pm}(\mathbf{k})/\partial t)_{\text{coll}} = & 2\pi/\hbar \cdot \sum_{\mathbf{k}'} [W_{\pm}(|\mathbf{k}-\mathbf{k}'|) f_1(\varepsilon_{\pm}) C(\varepsilon_{\pm}) (k_x' - k_x) \delta(E_k^{\pm} - E_{k'}^{\pm}) \\ & + V_{\mp}(|\mathbf{k}-\mathbf{k}'|) f_0(\varepsilon_{\pm}) f_0(-\varepsilon_{\pm} \mp x) \{k_x' C_{\mp}(\varepsilon_{\pm} \pm x) \\ & - k_x C_{\pm}(\varepsilon_{\pm})\} \delta(E_k^{\pm} - E_{k'}^{\mp} \pm (H_0 - 2H_c))]. \end{aligned} \quad (16)$$

Then, the Bloch equations are solved easily and give the following results

$$C_{\pm}(\varepsilon) = \phi_{\pm}^0(\varepsilon) (e\mathbf{F} - \nabla\zeta) - \phi_{\pm}^1(\varepsilon) \kappa \nabla T, \quad (17)$$

where

$$\phi_{\pm}^{\mu}(\varepsilon) = \frac{\pi \hbar^4}{m^2 \kappa T} \frac{\varepsilon^{\mu} A_{\mp}(\varepsilon \pm x) + (\varepsilon \pm x)^{\mu} B_{\pm}(\varepsilon)}{A_{\pm}(\varepsilon) A_{\mp}(\varepsilon \pm x) - B_{\pm}(\varepsilon) B_{\mp}(\varepsilon \pm x)}, \quad \mu=0, 1, \quad (18)$$

$$A_{\pm}(\varepsilon) = \frac{1}{4k_{\pm}^3} \int_0^{2k_{\pm}} W_{\pm}(q) q^3 dq + \frac{1}{2k_{\pm}} \frac{1+e^{-\varepsilon}}{1+e^{-\varepsilon \mp x}} \int_{\eta_{\pm}}^{\xi_{\pm}} V_{\mp}(q) q dq, \quad (19)$$

$$B_{\pm}(\varepsilon) = \frac{1}{2k_{\pm}} \frac{1+e^{-\varepsilon}}{1+e^{-\varepsilon \mp x}} \int_{\eta_{\pm}}^{\xi_{\pm}} V_{\mp}(q) \left\{ 1 - \frac{q^2}{2k_{\pm}^2} \mp \frac{m(2H_c - H_0)}{\hbar^2 k_{\pm}^2} \right\} q dq, \quad (20)$$

and

$$k_{\pm}^2 = \frac{2m}{\hbar^2} (E \pm H_c) = k_0^2 \left( 1 + \frac{\kappa T}{\zeta} \varepsilon \pm \frac{H_c}{\zeta} \right), \quad (21)$$

$$\xi_{\pm} = k_{\pm} \left\{ 1 + \left( 1 \mp \frac{2H_c - H_0}{E \pm H_c} \right)^{1/2} \right\}, \quad (22)$$

$$\eta_{\pm} = k_{\pm} \left| 1 - \left( 1 \mp \frac{2H_c - H_0}{E \pm H_c} \right)^{1/2} \right|. \quad (23)$$

### § 3. Results

The electric current  $j_x$  and the thermal flow  $Q_x$  are given by

$$j_x/e = L_{00}(eF - \nabla \zeta) - L_{01} \kappa \nabla T, \quad (24)$$

$$Q_x/\kappa T = L_{10}(eF - \nabla \zeta) - L_{11} \kappa \nabla T, \quad (25)$$

where

$$L_{\nu\mu} = \frac{1}{6\pi^2} \frac{\kappa T}{\hbar} \int_{-\infty}^{\infty} \varepsilon^{\nu} (k_{\pm}^3 \phi_{\pm}^{\mu}) f_1(\varepsilon) d\varepsilon, \quad (26)$$

and

$$(k_{\pm}^3 \phi_{\pm}^{\mu}) \equiv k_+^3 \phi_+^{\mu}(\varepsilon) + k_-^3 \phi_-^{\mu}(\varepsilon). \quad (27)$$

It is easily seen that Onsager's relations,  $L_{\mu\nu} = L_{\nu\mu}$ , is satisfied.

Using the conductivity tensor  $\mathbf{L}$ , we can calculate the various transport coefficients. But since the formula of  $\mathbf{L}$  is still very complicated, we use here the following approximations.

(1) We assume that  $W_{\pm}(q)$  and  $V_{\pm}(q)$  do not depend on  $q$  in the range  $0 < q < 2k_0$ , where  $k_0$  is the effective wave vector at Fermi surface determined by

$$\zeta = \hbar^2 k_0^2 / 2m. \quad (28)$$

(2) Because  $J/\zeta$ ,  $H_c/\zeta$ ,  $H_0/H_c$  and  $\kappa T/H_c$ , and furthermore  $J/A$  in dilute alloys, are sufficiently smaller than unity, we adopt only the leading terms concerning these factors.

Then  $\mathbf{L}$  is written as

$$L_{00} = \begin{cases} \frac{2}{3\pi} \frac{\hbar\zeta}{m} \int_{-\infty}^{\infty} \frac{f_1(\varepsilon) d\varepsilon}{W_0 + VF(\varepsilon)}, & \text{in metals} \\ \frac{2}{3\pi} \frac{\hbar\zeta}{m} \frac{1}{W_0} \left\{ 1 + \frac{W_1}{W_0} \frac{H_c}{\zeta} - \frac{V}{W_0} \frac{x}{1-e^{-x}} \right\}, & \text{in dilute alloys} \end{cases} \quad (29)$$

$$L_{01} = L_{10} = \begin{cases} \frac{2}{3\pi} \frac{\hbar\zeta}{m} \int_{-\infty}^{\infty} \frac{\varepsilon f_1(\varepsilon) d\varepsilon}{W_0 + VF(\varepsilon)} \left\{ \frac{\kappa T}{\zeta} \varepsilon + \frac{H_c}{\zeta} \frac{W_0 + 2VF(\varepsilon)}{W_0 + VF(\varepsilon)} \right\}, & \text{in metals} \\ \frac{2\pi}{9} \frac{\hbar\zeta}{m} \frac{1}{W_0} \left\{ \frac{\kappa T}{\zeta} - \frac{3}{\pi^2} \frac{W_1 V}{W_0} \frac{x^2}{1-e^{-x}} \right\}, & \text{in dilute alloys} \end{cases} \quad (30)$$

$$L_{11} = \begin{cases} \frac{2}{3\pi} \frac{\hbar\zeta}{m} \int_{-\infty}^{\infty} \frac{\varepsilon^2 f_1(\varepsilon) d\varepsilon}{W_0 + VF(\varepsilon)}, & \text{in metals} \\ \frac{2\pi}{9} \frac{\hbar\zeta}{m} \frac{1}{W_0} \left\{ 1 + \frac{W_1}{W_0} \frac{H_c}{\zeta} - \frac{V}{W_0} \frac{x + x^3/\pi^2}{1-e^{-x}} \right\}, & \text{in dilute alloys} \end{cases} \quad (31)$$

where

$$W_0 = \begin{cases} N^{-1}(g-1)^2 J^2(0) \{ \bar{j}_z^2 - (\bar{j})_z^2 \}, & \text{in metals} \\ N_i/N^2 \cdot \{ A^2(0) + (g-1)^2 J^2(0) \bar{j}_z^2 \}, & \text{in dilute alloys} \end{cases} \quad (32)$$

$$W_1 = N_i/N^2 \cdot 2(g-1) A(0) J(0) \bar{j}_z, \quad \text{in dilute alloys} \quad (33)$$

$$V = N_i/N^2 \cdot (g-1)^2 J^2(0) \{ j^2 + j - \bar{j}_z^2 - \bar{j}_z \}, \quad (34)$$

$$F(\varepsilon) = (1 + e^{-\varepsilon}) / (1 + e^{-\varepsilon - x}), \quad (35)$$

and  $\bar{j}_z$ ,  $\bar{j}_z^2$ , etc., are given by

$$\bar{j}_z = j + \frac{2j+1}{e^{(2j+1)x} - 1} - \frac{1}{e^x - 1}, \quad (36)$$

$$\bar{j}_z^2 = j^2 - \frac{2j-1}{e^x - 1} + \frac{2}{(e^x - 1)^2} - \frac{2j+1}{e^{(2j+1)x} - 1} - \frac{2(2j+1)}{(e^x - 1)(e^{(2j+1)x} - 1)}, \quad (37)$$

$$\bar{j}_z^2 - (\bar{j}_z)^2 = \frac{1}{(e^x - 1)(1 - e^{-x})} - \frac{(2j+1)^2}{(e^{(2j+1)x} - 1)(1 - e^{-(2j+1)x})}, \quad (38)$$

$$j^2 + j - \bar{j}_z^2 - \bar{j}_z = \frac{2}{e^x - 1} \left\{ j + \frac{2j+1}{e^{(2j+1)x} - 1} - \frac{1}{e^x - 1} \right\}. \quad (39)$$

(1) Electrical conductivity  $\sigma$ .

$\sigma$  is given by

$$\sigma = e^2 L_{00}, \quad (40)$$

and  $L_{00}$  is given by Eq. (29). The formula of  $L_{00}$  differs from that of the earlier papers<sup>4)5)</sup> because the degrees of the approximation to solve the Bloch equations are not equal. Nevertheless, the main character that  $\sigma$  increases with the increasing

magnetic order, that is the increase of  $x$ , is of course equal. The electrical resistivity at absolute zero temperature and in the state of no magnetic order are given by

$$(\sigma^{-1})_{T=0} = \begin{cases} 0 & \text{in metals} \\ \frac{3\pi}{2} \frac{m}{\hbar\zeta} \frac{N_i}{N^2 e^2} \left\{ A^2(0) + (g-1)^2 j^2 J^2(0) - 2(g-1)jJ(0)A(0) \frac{H_c}{\zeta} \right\}, & \text{in dilute alloys,} \end{cases} \quad (41)$$

and

$$(\sigma^{-1})_{x=0} = \begin{cases} \frac{3\pi}{2} \frac{m}{\hbar\zeta} \frac{1}{Ne^2} (g-1)^2 j(j+1) J^2(0), & \text{in metals,} \\ \frac{3\pi}{2} \frac{m}{\hbar\zeta} \frac{N_i}{N^2 e^2} \{ A^2(0) + (g-1)^2 j(j+1) J^2(0) \}, & \text{in dilute alloys.} \end{cases} \quad (42)$$

It is also easily seen from the above mentioned character that the electrical resistivity decreases when the external magnetic field is applied because  $x$  increases by the application of the external magnetic field.

In metals, if  $F(\varepsilon)$  is replaced approximately by unity, the result of [I] is obtained, namely

$$\sigma^{-1} = \frac{3\pi}{2} \frac{m}{Ne^2} \frac{(g-1)^2 J^2(0)}{\hbar\zeta} (j^2 + j - \bar{j}_z^2 - \bar{j}_z), \quad \text{in metals.} \quad (43)$$

(2) Thermal conductivity  $\kappa_0$  and Lorentz number  $L$ .  
 $\kappa_0$  and  $L$  are given by

$$\kappa_0 = \kappa^2 T \frac{L_{11}L_{00} - L_{10}L_{01}}{L_{00}}, \quad (44)$$

$$L = \frac{\kappa_0}{\sigma T} = \frac{\kappa^2}{e^2} \frac{L_{11}L_{00} - L_{10}L_{01}}{L_{00}^2}. \quad (45)$$

As is seen in Eqs. (29), (30) and (31),  $L_{10}$  and  $L_{01}$  are much smaller than  $L_{00}$  and  $L_{11}$  and thus we can write

$$L = \begin{cases} \frac{\kappa^2}{e^2} \frac{\int \varepsilon^2 f_1(\varepsilon) d\varepsilon}{W_0 + VF(\varepsilon)} \bigg/ \int \frac{f_1(\varepsilon) d\varepsilon}{W_0 + VF(\varepsilon)}, & \text{in metals,} \\ \frac{\pi^2}{3} \frac{\kappa^2}{e^2} \left\{ 1 - \frac{(g-1)^2}{\pi^2} \frac{J^2(0)}{A^2(0)} \frac{x^3}{1-e^{-x}} (j^2 + j - \bar{j}_z^2 - \bar{j}_z) \right\}, & \text{in dilute alloys.} \end{cases} \quad (46)$$

In metals  $L$  has a nearly constant value  $(\pi^2/3)(\kappa/e)^2$  over the whole range of  $x$ . This result is caused by the assumption of the isotropic scattering. As is seen in later section, if this assumption is not adopted  $L$  changes markedly from the value  $(\pi^2/3)(\kappa/e)^2$ .

(3) Absolute thermoelectric power  $\mathfrak{S}$ .

$\mathfrak{S}$  is given by

$$\mathfrak{S} = \frac{\kappa}{e} \frac{L_{01}}{L_{00}}. \quad (47)$$

In metals, neglecting  $W_0$  compared to  $VF(\varepsilon)$ , we obtain (see Appendix I) the following simplified approximated expression,

$$\mathfrak{S} = \frac{\kappa}{e} \left\{ \frac{\pi^2}{3} \frac{\kappa T}{\zeta} + 2 \frac{H_c}{\zeta} \frac{1 - e^{-x}}{1 + e^{-x}} \right\}. \quad (48)$$

The first term corresponds to the ordinary mechanism, namely this is characteristic of the isotropic scattering. The second term is characteristic of the *s-d* interaction and it is noticeable that this term is dominant than the first in sufficiently low temperature and remains to be of the finite value in absolute zero temperature. However, in general, there exists a some kind of lattice imperfection and thus  $W_0$  remains the finite value  $B_0$ . Then in sufficiently low temperature  $B_0$  becomes larger than  $VF(\varepsilon)$  and  $\mathfrak{S}$  is written as

$$\mathfrak{S} = \frac{\kappa}{e} \left\{ \frac{\pi^2}{3} \frac{\kappa T}{\zeta} + \frac{V^2}{B_0^2} \frac{H_c}{\zeta} x e^x \right\}. \quad (49)$$

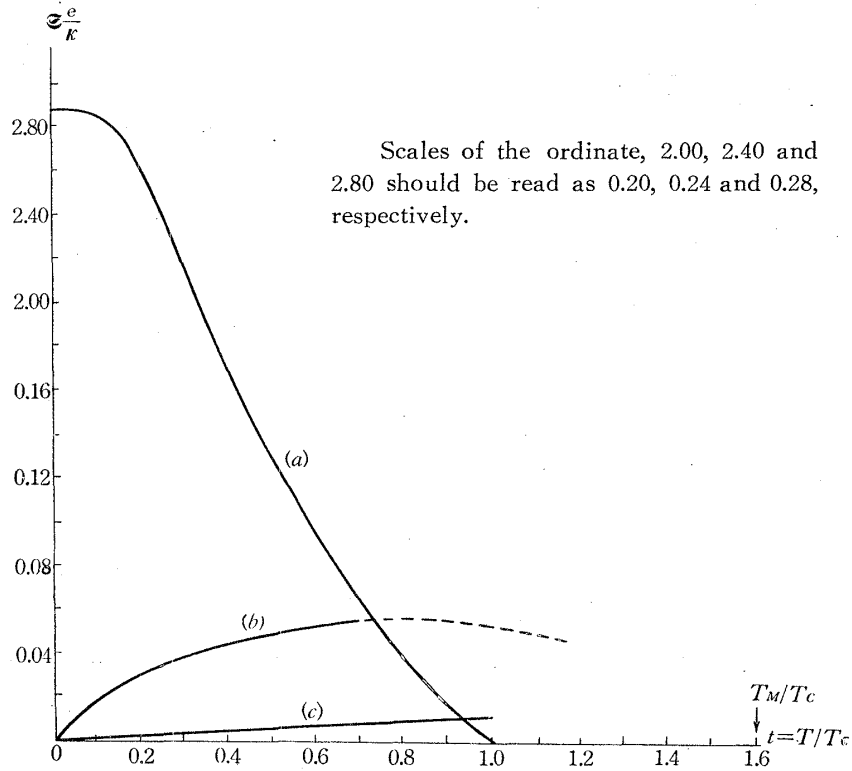


Fig. 1 The graph of  $\mathfrak{S}(e/\kappa)$  versus  $T/T_c$  (Gd)

- (a) means  $\mathfrak{S}_{\text{ano}}^{\text{mol}}$ , namely the anomalous part of  $\mathfrak{S}$  calculated by simple molecular field approximation.
- (b) means  $\mathfrak{S}_{\text{ano}}^{\text{spin}}$ , namely the anomalous part of  $\mathfrak{S}$  calculated by spin wave approximation.
- (c) means  $\mathfrak{S}_{\text{nor}}$ , namely the normal part of  $\mathfrak{S}$  and is given by  $0.011 t$ .

The temperature range where the above equation is applicable corresponds to that where the electrical resistance is nearly determined by the residual resistance. In Eq. (45) it is noticeable that the second term is proportional to  $(V/B_0)^2$  and the term proportional to  $V/B_0$  vanishes. This result is owing to the assumption that  $J(q)$  does not depend on  $q$ . In general, if  $J(q)$  depends on  $q$  the term proportional to  $(V/B_0)$  exists.

In dilute alloys,  $\mathcal{S}$  is written as

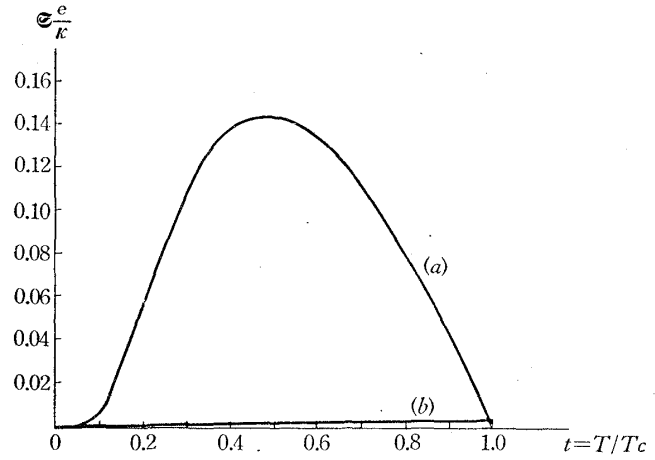


Fig. 2. The graph of  $\mathcal{S}(e/k)$  versus  $T/T_c$  (the dilute alloy of 1.8% Cu-Mn)

(a) means the anomalous part of  $\mathcal{S}$ .

(b) means the normal part of  $\mathcal{S}$  and is given by  $0.0008 t$ .

$$\mathcal{S} = \frac{\kappa}{e} \left\{ \frac{\pi^2}{3} \frac{\kappa T}{\zeta} - \frac{2(g-1)^3 J^3(0)}{A^3(0)} \bar{J}_z (j^2 + j - \bar{J}_z^2 - \bar{J}_z) \frac{x^2}{1 - e^{-x}} \right\}. \quad (50)$$

As mentioned above, if  $J(q)$  depends on  $q$  the term proportional to  $V/W_0$  should appear. For example, if we assume that  $J(q)$  is written by

$$J^2(q) = J^2(0) - J_1^2 q^2 / k_0^2, \quad (51)$$

the following term appears.

$$- \frac{\kappa}{e} \frac{1}{3} \frac{H_c}{\zeta} \frac{(g-1)^2 J_1^2}{A^2(0)} (j^2 + j - \bar{J}_z^2 - \bar{J}_z) \frac{x^2}{1 - e^{-x}}. \quad (52)$$

This term is nearly of the same order of magnitude as the second term of Eq. (50).

The more detailed comparison between the normal term and the anomalous terms are given in Figs. 1 and 2. (see § 5)

#### § 4. Calculations by spin wave approximation

In the preceding sections we calculated transport coefficients using the simplest molecular field model. But in sufficiently low temperatures, the magnetic ordered state may be represented by some kind of collective mode. In dilute alloys, however, it is a very complicated problem to treat the ordered state on the collective description. On the other hand, in metals the collective description of the ordered state is possible as was done in the previous paper.<sup>2)</sup> Therefore we treat in this section only the metallic ferromagnetics such as rare earth metals.

To perform the calculation we use the following model. (i) The magnetic spin system is always in the local thermal equilibrium. The justification of this



assumption may be fairly complicated because in a rigorous treatment the phonon system should be also considered, and then the so-called Umklapp process becomes important. (2) We assume here that the collective mode of the spin system is represented by the so-called spin wave mode and that the energy spectrum of the spin wave with wave vector  $\mathbf{k}$  is written by  $\varepsilon(\mathbf{k})$ .

(iii) Because we consider only sufficiently low temperature range, we adopt only the process in which only the one spin wave quantum is emitted or absorbed.

(iv) The square of the transition matrix element is given by

$$V_{\pm}(q) = \frac{2j(g-1)^2}{N} |J(q)|^2 \begin{bmatrix} N(q)+1 \\ N(q) \end{bmatrix}, \quad (53)$$

where  $\pm$  correspond respectively to the emission and the absorption of the spin wave, and  $N(q)$  means the distribution function of the spin wave with wave vector  $\mathbf{q}$  in the thermal equilibrium, namely

$$N(q) = \left[ \exp \frac{\varepsilon(q)}{\kappa T} - 1 \right]^{-1}. \quad (54)$$

Then we can write the Bloch equations concerning the electrons of  $\pm$  spin directions. The drift terms are same as Eq. (10). In collision terms of Eq. (11), the first term proportional to  $W_{\pm}(q)$  vanishes because it represents two spin wave processes, and only the last two terms are available substituting Eq. (53) for  $V_{\pm}(q)$  and  $\varepsilon(q)$  for  $H_0$ . Now, however, because it is impossible to solve these Bloch equations rigorously, we use here the technique of the total balance method.<sup>7)</sup> The assumptions adopted for the following calculation are as follows. (v) We adopt only the lowest terms concerning  $H_0/\zeta$  and  $\kappa T/\zeta$ . (vi) In Eq. (14) we assume that  $C_{\pm}(\varepsilon)$  is approximately written as

$$C_{\pm}(\varepsilon) = C_{\pm}^0 + C_{\pm}^1 \varepsilon. \quad (55)$$

Then the total balance method means that the Bloch equations are multiplied respectively by  $k_x$  and  $\varepsilon_{\pm} k_x$  and then integrated by  $d\mathbf{k}$ . These equations mean respectively that the change of the total value of  $k_x$  and  $\varepsilon_{\pm} k_x$  (or the total current and thermal flow) by drift effect and collision mechanisms are balanced in steady state, and from these four equations we can determine  $C_{\pm}^0$  and  $C_{\pm}^1$ . It is easily shown that in the case of electron-phonon interaction this method gives the same result as that of the second order variational method obtained by Sondheimer and Wilson.<sup>7)(8)</sup> (see Appendix II). Here we omit the detailed process of the calculation (see Appendix II) and give only the final results. Rewriting  $C_{\pm}^{\nu}$  such as

$$C_{\pm}^{\nu} = \phi_{\pm}^{\nu 0} (eF - \tau \zeta) - \phi_{\pm}^{\nu 1} \kappa T, \quad \nu = 0, 1, \quad (56)$$

conductivity tensor  $\mathbf{L}$  in Eq. (26) is obtained as follows,

$$L_{00} = \frac{1}{6\pi^2} \frac{\kappa T}{\hbar} (\phi_{\pm}^{00} k_{\pm 0}^3), \quad (57)$$

$$L_{11} = \frac{1}{18} \frac{\kappa T}{\hbar} (\phi_{\pm}^{11} k_{\pm 0}^3), \quad (58)$$

$$L_{01} = \frac{1}{6\pi^2} \frac{\kappa T}{\hbar} \left\{ (\phi_{\pm}^{01} k_{\pm 0}^3) + \frac{\pi^2}{2} \frac{\kappa T}{\zeta} (\phi_{\pm}^{11} k_{\pm 0}^3) \right\}, \quad (59)$$

$$L_{10} = \frac{1}{18} \frac{\kappa T}{\hbar} \left\{ \frac{3}{2} \frac{\kappa T}{\zeta} (\phi_{\pm}^{00} k_{\pm 0}^3) + (\phi_{\pm}^{10} k_{\pm 0}^3) \right\}, \quad (60)$$

where

$$(\phi_{\pm}^{\nu\mu} k_{\pm 0}^3) = k_{+0}^3 \phi_{+}^{\nu\mu} + k_{-0}^3 \phi_{-}^{\nu\mu} \quad (61)$$

$$k_{\pm 0}^2 = k_0^2 \left( 1 \pm \frac{H_c}{\zeta} \right), \quad (62)$$

and adopting only the leading terms with respect to  $\kappa T/\epsilon(q_0)$ , where  $q_0$  means the maximum wave vector of spin wave mode, we obtain

$$(\phi_{\pm}^{00} k_{\pm 0}^3) = \frac{2k_0^6}{[q^2 x]}, \quad (63)$$

$$(\phi_{\pm}^{11} k_{\pm 0}^3) = \frac{2\pi^2}{3} k_0^4 \frac{[x]}{[x][x^3] - [x^2]^2}, \quad (64)$$

$$\begin{aligned} (\phi_{\pm}^{01} k_{\pm 0}^3) &= \frac{2\pi^2}{3} k_0^6 \frac{\kappa T}{\zeta} \frac{1}{[q^2 x]} \\ &+ \frac{\pi^2}{3} k_0^4 \frac{H_c}{\zeta} \left( \frac{[x^2]}{[x][x^3] - [x^2]^2} + 3 \frac{[q^2 x^2]}{[q^2 x]} \frac{1}{4\pi[x] + [x^3]} \right), \end{aligned} \quad (65)$$

where  $[f(q, x)]$  such as  $[q^2 x]$  means the following value,

$$[f(q, x)] = \frac{1}{4\pi} \frac{m^2 \kappa T}{\hbar^4} \frac{2(g-1)^2 j}{N} \int_{k_0 H_c / \zeta}^{q_0} \frac{f(q, x) q |J(q)|^2}{(e^x - 1)(1 - e^{-x})} dq. \quad (66)$$

For example, we calculate transport coefficients in some simple cases.

Case A.  $\epsilon(q) = \epsilon_0 q/q_0$ ,  $J(q) = J_0$ .

In this case  $[f(q, x)]$  is written as

$$[f(q, x)] = \frac{1}{2\pi} \frac{(g-1)^2 j J_0^2}{N} \frac{m^2 \kappa T}{\hbar^4} q_0^2 t^2 \int_{x_0}^{t^{-1}} \frac{x f(q_0 t x, x)}{(e^x - 1)(1 - e^{-x})} dx, \quad (67)$$

where

$$t = \kappa T / \epsilon_0, \quad x_0 = \frac{1}{t} \frac{H_c}{\zeta} \frac{k_0}{q_0}, \quad (68)$$

and from Eqs. (40), (57) and (63) we obtain

$$\sigma = \frac{4}{3\pi} \frac{Ne^2}{m} \frac{\hbar \zeta}{j(g-1)^2 J_0^2} \left( \frac{k_0}{q_0} \right)^4 \frac{1}{t^4} \frac{1}{\phi_4(t^{-1}, x_0)}, \quad (69)$$

$$L = \frac{\kappa^2}{e^2} \frac{\pi^4}{9} \left( \frac{q_0}{k_0} \right)^2 t^2 \left\{ 1 - \frac{\phi_3^2(t^{-1}, x_0)}{\phi_2(t^{-1}, x_0) \phi_4(t^{-1}, x_0)} \right\}, \quad (70)$$

$$\begin{aligned} \mathfrak{S} = \frac{\kappa}{e} \frac{\pi^2}{3} \frac{\kappa T}{\zeta} \left\{ 1 + \frac{3}{2} \left( \frac{q_0}{k_0} \right)^2 \frac{H_e}{\epsilon_0} t \left( \frac{1}{3} \frac{\phi_3(t^{-1}, x_0) \phi_4(t^{-1}, x_0)}{\phi_2(t^{-1}, x_0) \phi_4(t^{-1}, x_0) - \phi_3(t^{-1}, x_0)^2} \right. \right. \\ \left. \left. + \frac{\phi_5(t^{-1}, x_0)}{4\pi^2 \phi_2(t^{-1}, x_0) + \phi_4(t^{-1}, x_0)} \right) \right\}, \quad (71) \end{aligned}$$

and where

$$\phi_n(t^{-1}, x_0) = \int_{x_0}^{t^{-1}} \frac{x^n dx}{(e^x - 1)(1 - e^{-x})}. \quad (72)$$

When  $x_0$  is sufficiently smaller and  $t^{-1}$  is sufficiently larger than unity, we can replace  $\phi_n(t^{-1}, x_0)$  by  $\phi_n(\infty, 0)$ . The values of  $\phi_n(\infty, 0)$  are given as follows,

$$\begin{aligned} \phi_2(\infty, 0) = 3.29, \quad \phi_3(\infty, 0) = 7.22 \\ \phi_4(\infty, 0) = 25.9, \quad \phi_5(\infty, 0) = 124.4. \end{aligned} \quad (73)$$

Then  $\sigma$  is proportional to  $T^{-4}$ ,  $L$  to  $T^2$  and  $\mathfrak{S}$  is written as

$$\mathfrak{S} = \frac{\kappa}{e} \frac{\pi^2}{3} \frac{\kappa T}{\zeta} \left\{ 1 + 4 \left( \frac{q_0}{k_0} \right)^2 \frac{H_e \kappa T}{\epsilon_0^2} \right\}. \quad (74)$$

When  $x_0$  is sufficiently larger than unity, that is, in very low temperature range, we can replace  $\phi_n(t^{-1}, x_0)$  by

$$\begin{aligned} \phi_n(t^{-1}, x_0) &\Rightarrow \int_{x_0}^{\infty} x^n e^{-x} dx \\ &= (x_0^n + n x_0^{n-1} + \dots + n!) e^{-x_0} \end{aligned} \quad (75)$$

and thus  $\sigma$  is proportional to  $e^{x_0}$ . On the otherhand, the denominators of  $L$  and  $\mathfrak{S}$  vanish if we take only the leading term of Eq. (75). Therefore we must calculate the values of  $L$  and  $\mathfrak{S}$  in such a very low temperature range using the full expressions as given in Appendix II. However, in such a very low temperature range, we must consider the following facts that (i) The approximation to replace  $C_{\pm}(\epsilon)$  by  $C_{\pm}^0 + C_{\pm}^1 \epsilon$  becomes very poor. (ii) The Raman process in which one spin wave is absorbed and the other one emitted becomes important because in this process there are no lower limit with respect to  $q$ . (iii) Furthermore, in actual case, the transport phenomena of such a low temperature range are determined mostly by residual resistance. The more detailed calculation by considering such a circumstance may be carried out in future.

Case B

$$\epsilon = \epsilon_0 q^2 / q_0^2, \quad J(q) = J_0.$$

In this case  $[f(q^2, x)]$  is written as

$$[f(q^2, x)] = \frac{1}{4\pi} \frac{j(g-1)^2 J^2(0)}{N} \frac{m^2 \kappa T}{\hbar^4} q_0^2 t \int_{x_0}^{t^{-1}} \frac{f(q_0^2 t x, x)}{(e^x - 1)(1 - e^{-x})} dx, \quad (76)$$

$$\text{where } t = \frac{\kappa T}{\epsilon_0}, \quad x_0 = \frac{1}{t} \left( \frac{H_c}{\zeta} - \frac{k_0}{q_0} \right)^2, \quad (77)$$

and thus we obtain

$$\sigma = \frac{8}{3\pi} \frac{Ne^2}{m} \frac{\hbar \zeta}{j(g-1)^2 J^2(0)} \left( \frac{k_0}{q_0} \right)^4 \frac{1}{t^2} \frac{1}{\phi_2(t^{-1}, x_0)}, \quad (78)$$

$$L = \frac{\kappa^2}{e^2} \frac{\pi^4}{9} \left( \frac{q_0}{k_0} \right)^2 t / \left\{ \frac{\phi_3(t^{-1}, x_0)}{\phi_2(t^{-1}, x_0)} - \frac{\phi_2(t^{-1}, x_0)}{\phi_1(t^{-1}, x_0)} \right\}, \quad (79)$$

$$\begin{aligned} \mathfrak{S} = & \frac{\kappa}{e} \frac{\pi^2}{3} \left[ 1 + \frac{1}{2} \left( \frac{q_0}{k_0} \right)^2 \frac{H_c}{\epsilon_0} \left\{ \left( \frac{\phi_1(t^{-1}, x_0) \phi_3(t^{-1}, x_0)}{\phi_2(t^{-1}, x_0)^2} - 1 \right)^{-1} \right. \right. \\ & \left. \left. + 3 \left( 4\pi^2 \frac{\phi_1(t^{-1}, x_0)}{\phi_3(t^{-1}, x_0)} + 1 \right)^{-1} \right\} \right] \frac{\kappa T}{\zeta}. \end{aligned} \quad (80)$$

When  $x_0$  is sufficiently smaller and  $t^{-1}$  larger than unity we can replace  $\phi_n(t^{-1}, x_0)$  by  $\phi_n(\infty, 0)$  as was shown in Eq. (73). However, this replacement is impossible for the function  $\phi_1(t^{-1}, x_0)$  because  $\phi_1(t^{-1}, x_0)$  diverges logarithmically in the limit of  $x_0 \rightarrow 0$ . Considering the above fact,  $\sigma$  is proportional to  $T^{-2}$  and  $L$  proportional to  $T$ . While  $\mathfrak{S}$  is written as

$$\mathfrak{S} = \frac{\kappa}{e} \frac{\pi^2}{3} \frac{\kappa T}{\zeta} \left\{ 1 + 1.5 \left( \frac{q_0}{k_0} \right)^2 \frac{H_c}{\epsilon_0} \frac{1}{\phi_1(t^{-1}, x_0)} \right\}, \quad (81)$$

and in this case, too, the anomalous term becomes large because the value of  $H_c/\epsilon_0$  is in general of the order of 10 and  $\ln x_0$  is not so large (see § 5).

In the case of antiferromagnetic ordering the same treatment is possible (see Appeneix II). For example, the explicit results of the case A are given as follows.

$$\sigma = \frac{2}{3\pi} \frac{Ne^2}{m} \frac{\hbar \zeta}{j(g-1)^2 J_0^2} \left( \frac{k_0}{q_0} \right)^4 \frac{1}{t^4} \frac{1}{\phi_4(t^{-1})}, \quad (82)$$

$$L = \frac{\kappa^2}{e^2} \frac{\pi^4}{9} \left( \frac{q_0}{k_0} \right)^2 t^2 / \left[ 1 + \frac{\pi^2}{3} \left( \frac{q_0}{k_0} \right)^2 t^2 \left\{ 1 - \frac{1}{2\pi^2} \phi_6(t^{-1}) / \phi_4(t^{-1}) \right\} \right], \quad (83)$$

$$\mathfrak{S} = \frac{\kappa}{e} \frac{\pi^2}{3} \frac{\kappa T}{\zeta} \frac{1 + \pi^2 (q_0/k_0)^2 t^2 \{ 1 - (1/4\pi^2) \phi_6(t^{-1}) / \phi_4(t^{-1}) \}}{1 + (\pi^2/3) (q_0/k_0)^2 t^2 \{ 1 - (1/2\pi^2) \phi_6(t^{-1}) / \phi_4(t^{-1}) \}}, \quad (84)$$

where

$$\phi_n(t^{-1}) = \int_0^{t^{-1}} \frac{x^n dx}{(e^x - 1)(1 - e^{-x})}, \quad (85)$$

and  $t = \kappa T / \epsilon_0$ .

In antiferromagnetic ordering,  $H_c$  becomes zero and thus the anomalous term of  $\mathfrak{S}$  proportional to  $H_c/\zeta$  disappears.

### § 5. Conclusions and discussions

(i) By *s-d* interaction, anomalous thermoelectric force as well as electrical and thermal resistivities appears both in ferromagnetic metals and alloys.

(ii) For the more qualitative discussions we take Gd as an example of ferromagnetic metals. Then using a free electron model we can obtain the following values.

$$\begin{aligned} n/N &= 3, & \zeta &= 1.2 \times 10^{-11} \text{ erg}, & J_0 &= 2.5 \times 10^{-13} \text{ erg}, \\ g &= 2, & j &= 7/2, \\ H_c &= 1.75 \times 10^{-12} \bar{J}_z / j \text{ erg}. \end{aligned} \quad (86)$$

If we assume that there is only the *s-d* exchange interaction,  $H_0$  is written as

$$H_0 = H_m \bar{J}_z, \quad (87)$$

and  $H_m$  is determined experimentally by the paramagnetic Curie temperature  $T_p$  such as

$$\kappa T_p = \frac{j(j+1)}{3} H_m \quad (88)$$

and thus in Gd we obtain

$$T_p = 300^\circ \text{ K}, \quad H_m = 8 \times 10^{-15} \text{ erg} = 57\kappa. \quad (89)$$

$\epsilon_0$  is also determined experimentally by  $T_M$  as follows,

$$\epsilon_0 = \left\{ \frac{3}{2j} \Gamma\left(\frac{3}{2}\right) \zeta\left(\frac{3}{2}\right) \right\}^{2/3} \kappa T_M = \kappa T_M, \quad (90)$$

where  $T_M$  is obtained as an effective Curie temperature by extrapolating the magnetization curve of the range where  $T^{3/2}$  law is applicable in a good approximation. In Gd we obtain the following results.

$$\begin{aligned} T_M &= 470^\circ \text{ K}, & \epsilon_0 &= 5.8 \times 10^{-14} \text{ erg}, \\ x_0 &= \frac{\epsilon_0}{\kappa T} \left( \frac{H_c}{\zeta} \frac{k_0}{q_0} \right)^2 = \frac{13}{T}, \end{aligned} \quad (91)$$

and the  $T^{3/2}$  law of the magnetization curve is applicable still up to  $200^\circ \text{ K}$ . In Fig. 1 the curve of  $\mathfrak{S}$  is plotted as a function of  $T$ . Experimental value of  $\mathfrak{S}$  is, however, not available.

(iii) As is well-known, the electrical resistivity by the ordinary electron phonon interaction, say  $\rho_{ep}$ , is proportional to  $T^5$  in temperature sufficiently lower than the Debye temperature. While the resistivity by *s-d* exchange interaction, say  $\rho_{sd}$ , is proportional to  $T^2$  from Eq. (78). Therefore the following behaviour of  $\rho$  is expected that in sufficiently low temperature  $\rho_{sd}$  overcomes  $\rho_{ep}$  and  $\rho$  is proportional to  $T^2$ . In Gd, however, the temperature dependence of  $\rho$  differs from

the expected behaviour. This result seems to be owing to the fact that in rare earth metals the temperature dependence of  $\rho_{ep}$  may be fairly different from that of the ordinary theory because of the complicated band structure and thus it is very difficult to separate  $\rho$  into  $\rho_{ep}$  and  $\rho_{sd}$  in good accuracy. For example, the electrical resistivity of La at sufficiently low temperature is rather proportional to  $T$ . Recently Mannari pointed out that in Fe  $\rho$  changes with  $T^2$  in low temperature.<sup>9)</sup> In the transition metals, too,  $\rho_{ep}$  seems to be fairly different from that of the ordinary theory as is seen in *Pd* and *W* and thus it is still ambiguous whether this temperature dependence is due to the *s-d* interaction or not.

(iv) In rare earth metals the thermal conductivity has not so far been measured. In the above calculations we neglected the thermal flow due to spin diffusion or spin wave flow. But it seems for me that the effect of spin diffusion may be important because of the long range character of the *s-d* interaction. The more detailed discussion about this point may be made in future.

(v) For an example of dilute alloys we take up here the Cu-Mn alloys as one of the simplest substances because  $\text{Mn}^{++}$  is in the state of  $^5S$  and has no orbital momentum. Notwithstanding of this circumstance the actual magnetic state of Cu-Mn alloys seems to be fairly complicated and the interactions other than the *s-d* interaction seems to be important for the magnetic ordering. However, with respect to the electrical and thermal resistivities the results of Eqs. (40) and (44) do not depend so sensitively on the character of the magnetic ordering and the agreement with the experimental results is fairly good as was shown by Yosida.<sup>5)</sup> While the anomalous part of  $\mathfrak{S}$  depends so sensitively on the character of the magnetic ordering. For example, if the magnetic spins are in a state of some kind of the antiferromagnetic ordering, the anomalous part of  $\mathfrak{S}$  disappears. However, on the other hand, for the applicability of Eq. (50) it is not necessary that there appears the resultant magnitude of the magnetization because Eq. (50) does not depend on the direction of  $\bar{j}_z$ . For example, Eq. (50) is applicable even in the case that the ferromagnetic ordering exists only in the limited range of the mean free path of the conduction electron's spin. Therefore the measurement of  $\mathfrak{S}$  may give a certain knowledge about the magnetic ordering of dilute alloys. For a more quantitative discussions we take up the sample of 1.8 atomic per cent  $\text{Mn}^{10)}$  which was chosen by Yosida for the discussion of the anomalous electrical resistivity. Then using the free electron model we obtain the following values.

$$\begin{aligned}
 (\sigma^{-1})_{T=0} &= 4.6\mu \text{ ohm} = 5.1 \times 10^{-18} \text{ e.s.u.}, \\
 (\sigma^{-1})_{x=0} &= 5\mu \text{ ohm} = 5.55 \times 10^{-18} \text{ e.s.u.}, \\
 g &= 2, \quad j = 5/2, \quad N_i/N = 0.018, \\
 \zeta &= 1.1 \times 10^{-11} \text{ erg.}, \quad N = 8.5 \times 10^{22} \text{ c.c}^{-1}, \\
 A(0) &= 3.4 \times 10^{-12} \text{ erg.}, \quad J(0) = 0.7 \times 10^{-12} \text{ erg.},
 \end{aligned} \tag{92}$$

$$H_c = 3.15 \times 10^{-14} \text{ erg.}, \quad T_p \sim 20^\circ \text{ K.}$$

From the above value it is easily seen that the term proportional to  $H_c/\zeta$  in Eq. (52) is smaller than that proportional to  $J(0)/A(0)$  and thus we can use Eq. (50) in a good approximation. The graph of  $\mathfrak{S}$  given by Eq. (50) is plotted in Fig. 2. The anomalous thermoelectric effect is very much larger than the normal part. For example, the maximum value of the anomalous part is 350 times of the normal part at  $10^\circ \text{K}$ .

Even in the case of non-ferromagnetic ordering, the anomalous part of  $\mathfrak{S}$  is observable by applying strong magnetic field. For example, using a simplest model, we can replace  $H_0$  by

$$H_0 = g\beta H, \quad (93)$$

and taking only the linear terms with respect to the external field  $H$ , we obtain

$$\mathfrak{S} = \frac{\kappa}{e} \left\{ \frac{\pi^2}{3} \frac{\kappa T}{\zeta} - \frac{4}{9} g^2 (g-1)^2 j^2 (j+1)^2 \left( \frac{J(0)}{A(0)} \right)^3 \left( \frac{\beta H}{\kappa T} \right)^2 \right\}. \quad (94)$$

For example, taking the above data of 1.8% Mn-Cu alloy, we obtain

$$\mathfrak{S} = \frac{\kappa}{e} \frac{\pi^2}{3} \frac{\kappa T}{\zeta} \left\{ 1 - 13 \frac{(10^{-4} H)^2}{(10^{-1} T)^3} \right\}, \quad (95)$$

and we can see that when the external field of  $10^4$  gauss is applied the anomalous part dominates the normal part in the temperature range lower than  $22^\circ \text{K}$ .

Measurement of  $\mathfrak{S}$  in various Cu alloys has been carried out so far<sup>11)</sup> and the anomalously large values of  $\mathfrak{S}$  were observed in samples which exhibit resistance anomalies. It seems to me that one of the mechanisms of such anomalous thermoelectric power may be what is considered in this paper. However, because  $\mathfrak{S}$  depends sensitively on the ordered states of impurity atoms or the band structure of Cu and these are considered as fairly complicated in Cu alloys (the sign of  $\mathfrak{S}$  in pure Cu is inverse from that predicted by the free electron model), the detailed qualitative comparison between our theoretical and experimental values is not tried here. Measurement in a strong magnetic field or in alkaline metals is desired.

(vi) The mechanism of anomalous thermoelectric power may be recognized as follows. When temperature gradient  $\nabla T$  exists, the electron distribution function  $f(\mathbf{k})$  changes from the thermal equilibrium values as shown in Fig. 3a. On the other hand, the inelastic scattering causes a mixing as shown in Fig. 3a and thus the stationary distribution function becomes such as shown in Fig. 3b. If there are no other different circumstances between the systems of  $\pm$  spin electron, the situation of Fig. 3b exhibits no anomalous thermoelectric current. However, if there are some differences between  $\pm$  spin electrons, for example, the difference of the transition probability of elastic scattering in dilute alloys or the existence of the effective field in metallic ferromagnetics, the anomalous thermoelectric current flows as shown in Fig. 3c and thus  $\mathfrak{S}_{ano}$  appears.

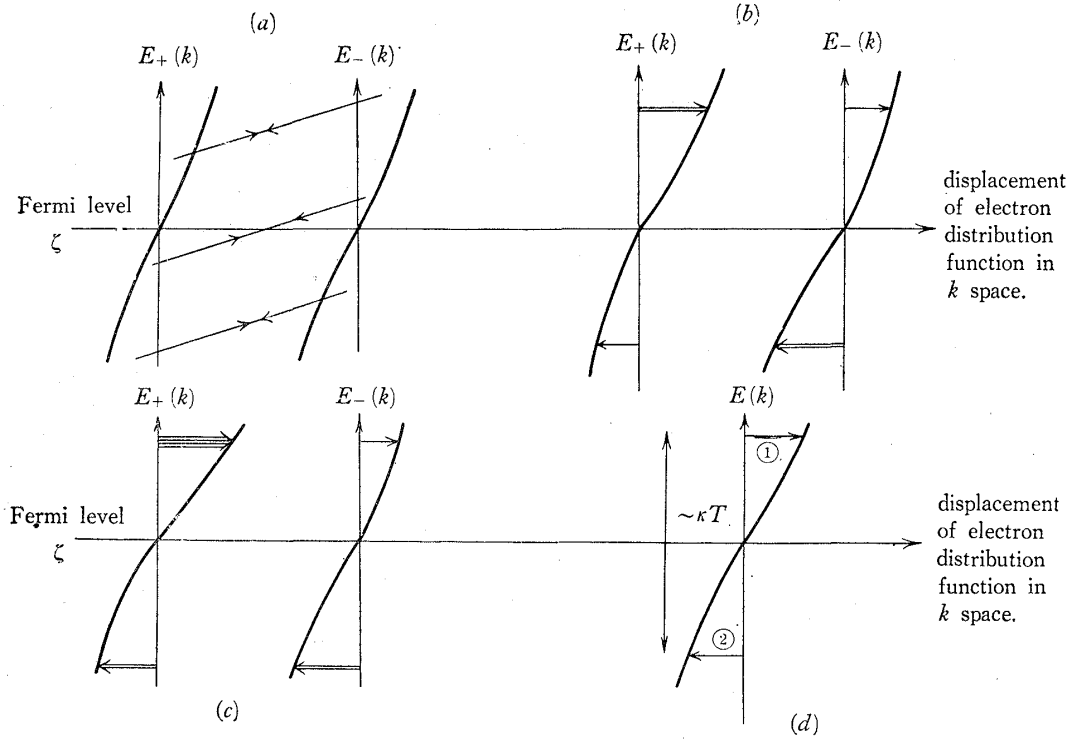


Fig. 3 Illustration of the mechanism of the anomalous thermoelectric power. (a), (b) and (c) show the mechanism of anomalous thermoelectric power. (d) shows the mechanism of normal thermoelectric power  $\mathcal{S}_{nor}$ .  $\mathcal{S}_{nor}$  is proportional to the difference of the displacements ① and ②, and thus usually of the order of  $\kappa T/\zeta$ .

### Appendix I

The integrals to be solved are such as

$$I = \int_{-\infty}^{\infty} \frac{1 + e^{-\varepsilon - x}}{(e^{\varepsilon} + 1)(1 + e^{-\varepsilon})^2} \varepsilon^n d\varepsilon, \quad n=0, 1, 2. \quad (\text{A1})$$

When  $n$  means an even number, we can write the integrand as follows,

$$\frac{\varepsilon^n}{(e^{\varepsilon} + 1)(1 + e^{-\varepsilon})} - (1 - e^{-x}) \frac{\varepsilon^n}{(e^{\varepsilon} + 1)^2(1 + e^{-\varepsilon})}. \quad (\text{A2})$$

The first integral is easily performable. The second integral is also performable using the following relation,

$$\frac{1}{(e^{\varepsilon} + 1)^2(1 + e^{-\varepsilon})} + \frac{1}{(e^{\varepsilon} + 1)(1 + e^{-\varepsilon})^2} = \frac{1}{(e^{\varepsilon} + 1)(1 + e^{-\varepsilon})}. \quad (\text{A3})$$

Thus  $I_n$  is obtained as follows,

$$\begin{aligned} I_0 &= 1/2 \cdot (1 + e^{-x}), \\ I_2 &= \pi^2/6 \cdot (1 + e^{-x}), \text{ etc.} \end{aligned} \quad (\text{A4})$$

When  $n$  means an odd number, we can calculate  $I_n$  as follows,



$$\begin{aligned}
I_n &= -1/2 \cdot (1 - e^{-x}) \int_{-\infty}^{\infty} \varepsilon^n \frac{d}{d\varepsilon} \left\{ \frac{1}{(e^\varepsilon + 1)(1 + e^{-\varepsilon})} \right\} d\varepsilon \\
&= \frac{n}{2} (1 - e^{-x}) \int_{-\infty}^{\infty} \frac{\varepsilon^{n-1}}{(e^\varepsilon + 1)(1 + e^{-\varepsilon})} d\varepsilon.
\end{aligned} \tag{A5}$$

Thus

$$I_1 = \frac{1}{2} (1 - e^{-x}). \tag{A6}$$

## Appendix II

The equations of the total balance with respect to  $k_x$  and  $\varepsilon_\pm k_x$  are written as

$$d_\pm^\nu = \mathcal{D}_\pm^\nu, \quad \nu = 0, 1 \tag{A7}$$

where

$$d_\pm^0 = \frac{1}{6\pi^2 \hbar} \left\{ k_{\pm 0}^3 (eF - \nu \zeta) - \frac{\pi^2}{2} \frac{\kappa T}{\zeta} k_0^3 \kappa \nu T \right\}, \tag{A8}$$

$$d_\pm^1 = \frac{1}{6\pi^2 \hbar} \left\{ \frac{\pi^2}{2} \frac{\kappa T}{\zeta} k_0^3 (eF - \nu \zeta) - \frac{\pi^2}{3} k_{\pm 0}^3 \kappa \nu T \right\}, \tag{A9}$$

$$\begin{aligned}
\mathcal{D}_\pm^\nu &= \frac{1}{12\pi^3} \frac{m^2 \kappa T}{\hbar^5} \frac{2j(g-1)^2}{N} \int_{-\infty}^{\infty} \varepsilon^\nu d\varepsilon \int_{k_0 H_c / \zeta}^{q_0} dq \frac{\pm q |J(q)|^2}{(e^{\varepsilon \pm x} + 1)(1 - e^{-\varepsilon})(1 - e^{\mp x})} \\
&\times \left[ \left( \frac{q^2}{2} - k_0^2 \mp k_0^2 \frac{x}{2} \frac{\kappa T}{\zeta} - k_0^2 \varepsilon \frac{\kappa T}{\zeta} \right) (C_\mp^0 + (\varepsilon \pm x) C_\mp^1) \right. \\
&\quad \left. + \left( k_{\pm 0}^2 + k_0^2 \varepsilon \frac{\kappa T}{\zeta} \right) (C_\pm^0 + \varepsilon C_\pm^1) \right].
\end{aligned} \tag{A10}$$

Because, as is seen from Eq. (61), the quantities we require are  $(k_{\pm 0}^3 \phi_\pm^{\nu\mu})$ , we transform  $\{C_\pm^\nu\}$  into  $\{\alpha_\nu, \beta_\nu\}$  as follows,

$$k_{+0}^3 C_+^\nu + k_{-0}^3 C_-^\nu \equiv (k_{\pm 0}^3 C_t^\nu) = \alpha_\nu, \tag{A11}$$

$$k_{+0}^3 C_+^\nu - k_{-0}^3 C_-^\nu = \beta_\nu. \tag{A12}$$

Then the matrix to determine  $\{\alpha_\nu, \beta_\nu\}$  is written by using the approximation to adopt only the leading terms with respect to  $\kappa T/\zeta$  and  $H_c/\zeta$  as follows.

$\alpha_0,$	$\beta_0,$	$\alpha_1,$	$\beta_1$	$\frac{eF - \nu \zeta}{\kappa \nu T}$
$\frac{q^2}{2} x,$			$-\frac{q^2}{4} x^2,$	$k_0^6$
	$-2x\mathcal{A} - \frac{x^2}{2}\delta,$	$x^2\mathcal{A} + \frac{x^3}{4}\delta,$		$-\frac{\pi^2}{2} \frac{\kappa T}{\zeta} k_0^6$
	$-\left(2k_0 - \frac{q^2}{2}\right)x,$	$\left(2k_0 - \frac{q^2}{2}\right)\frac{x^2}{2},$		$0$

$$\begin{array}{ccc}
2x\Delta + \frac{x^2}{2}\delta, & -x^2\Delta - \frac{5x^3+8\pi^2x}{12}\delta, & 0 \\
\hline
- \left(2k_0 - \frac{q^2}{2}\right) \frac{x^2}{2}, & \frac{1}{6} \left\{ 3k_0^2 x^3 + q^2 \left( \pi^2 x - \frac{x^3}{2} \right) \right\}, & \frac{\pi^2}{2} \frac{\kappa T}{\zeta} k_0^6 \\
\hline
x^2\Delta + \frac{x^3}{4}\delta, & -\frac{2}{3}(x^3 + \pi^2 x)\Delta - \frac{x^4 + 2\pi^2 x}{4}\delta, & -\frac{\pi^2}{3} k_0^6 \\
\hline
\frac{q^2 x^2}{4}, & -\frac{1}{6} \left\{ k_0^2 (x^3 + 4\pi^2 x) - q^2 \left( \pi^2 x - \frac{x^3}{2} \right) \right\}, & 0 \\
\hline
-x^2\Delta - \frac{5x^3+8\pi^2x}{12}\delta, & \frac{2}{3}(x^3 + \pi^2 x)\Delta + \frac{x^4 + 2\pi^2 x^2}{4}\delta, & 0 \\
\hline
\end{array}
\tag{A13}$$

where

$$\Delta = k_0^2 \frac{H_c}{\zeta}, \quad \delta = k_0^2 \frac{\kappa T}{\zeta}, \tag{A14}$$

and  $f(q, x)$  means (in Eq. (66) it is written as  $[f(q, x)]$ )

$$\frac{1}{2\pi} \frac{m^2 \kappa T}{\hbar^4} \frac{j(g-1)^2}{N} \int_{k_0 H_c / \zeta}^{q_0} \frac{f(q, x) q |J^2(q)|^2}{(e^x - 1)(1 - e^{-x})} dq. \tag{A15}$$

From the above matrix, we can easily obtain the required quantities as follows.

$$(i) \quad (\phi_{\pm}^{00} k_{\pm 0}^3) = k_0^6 \frac{-k_0^2(4\pi^2 x + x^3) + q^2(\pi^2 x - x^3/2)}{D_a}, \tag{A16}$$

where

$$D_a = \begin{vmatrix} \frac{q^2 x}{2}, & -\frac{q^2 x^2}{4} \\ \frac{q^2 x^2}{4}, & -\frac{k_0^2(4\pi^2 x + x^3) - q^2(\pi^2 x - x^3/2)}{6} \end{vmatrix}. \tag{A17}$$

Because the available wave vector  $q$  of the spin wave is much smaller than  $k_0$  in low temperature, we pick up only the leading term with respect to  $q/k_0$ . Then we obtain

$$(\phi_{\pm}^{00} k_{\pm 0}^3) = \frac{2k_0^6}{q^2 x}. \tag{A18}$$

$$(ii) \quad (\phi_{\pm}^{11} k_{\pm 0}^3) = \frac{\pi^2}{3} k_0^6 \frac{(2k_0^2 - q^2/2)x}{D_b}, \tag{A19}$$

where

$$D_b = \begin{vmatrix} -\left(2k_0^2 - \frac{q^2}{2}\right)x, & \left(2k_0^2 - \frac{q^2}{2}\right)\frac{x^2}{2} \\ -\left(2k_0^2 - \frac{q^2}{2}\right)\frac{x^2}{2}, & \frac{1}{2}k_0^2x^3 - \frac{1}{6}q^2(\pi^2x - x^3/2) \end{vmatrix}, \quad (\text{A20})$$

and taking only the leading terms respecting  $q/k_0$  we obtain

$$(\phi_{\pm}^{11} k_{\pm 0}^3) = \frac{2\pi^2}{3} k_0^4 \frac{[x]}{[x][x^3] - [x^2]^2}. \quad (\text{A21})$$

$$\begin{aligned} (\text{iii}) \quad (\phi_{\pm}^{01} k_{\pm 0}^3) &= \frac{\pi^2}{2} \frac{\kappa T}{\zeta} (\phi_{\pm}^{00} k_{\pm 0}^3) \\ &+ \frac{\pi^2}{3} \frac{k_0^6}{D_a D_b} \begin{vmatrix} -2x\Delta - \frac{x^2}{2}\delta, & x^2\Delta + \frac{x^3}{4}\delta, & -\frac{q^2x^2}{4} \\ -\left(2k_0^2 - \frac{q^2}{2}\right)x, & \left(2k_0^2 - \frac{q^2}{2}\right)\frac{x^2}{2}, & 0 \\ -x^2\Delta - \frac{5x^3 + 8\pi^2x}{12}\delta, & \frac{2}{3}(x^3 + \pi^2x)\Delta + \frac{x^4 + 2\pi^2x^2}{4}\delta, & \\ & \frac{-k_0^2(4\pi^2x + x^3) + q^2(\pi^2x - x^3/2)}{6} \end{vmatrix}, \end{aligned} \quad (\text{A22})$$

and taking only the leading terms with respect to  $q/k_0$ , we obtain the result of Eq. (65).

If one wishes to obtain the ordinary results of electron-phonon interaction, the following simplification should be made. (i) The behaviours of the electron systems with  $\pm$  spin directions are equivalent and thus in the matrix of Eq. (A13)  $\beta_0$  and  $\beta_1$  should be put zero and the second and fourth lines of the matrix be omitted. (ii) The effect of the scattering mechanisms becomes twice that in the above case because in the present case the two mechanisms corresponding to the emission and absorption of phonons are available. (iii) The transition probability given by Eq. (53) should be replaced by the ordinary value of electron-phonon interaction. Then the matrix to determine  $\alpha_0$  and  $\alpha_1$  becomes such as

$\alpha_0$	$\alpha_1$	$eF - p\zeta$	$\kappa T$
$q^2x,$	$k_0^2 \frac{x^3}{2} \frac{\kappa T}{\zeta},$	$k_0^6$	$-\frac{\pi^2}{2} \frac{\kappa T}{\zeta} k_0^6$
$k_0^2 \frac{x^3}{2} \frac{\kappa T}{\zeta},$	$\frac{1}{3} \left\{ 3k_0^2x^3 + q^2(\pi^2x - \frac{x^3}{2}) \right\},$	$\frac{\pi^2}{2} \frac{\kappa T}{\zeta} k_0^6,$	$-\frac{\pi^2}{3} k_0^6$

(A23)

and this gives just the same result as that calculated by Sondheimer and Wilson using the second order variational method.

In the case of the antiferromagnetic ordering, the situation is just the same as that of the electron phonon interaction and the matrix of (A23) is also applicable. The explicit forms of  $(\phi_{\pm}^{\nu\mu} k_{\pm 0}^3)$  are given as follows.

$$(\phi_{\pm}^{00} k_{\pm 0}^3) = \frac{k_0^6}{q^2 x}, \quad (\text{A24})$$

$$(\phi_{\pm}^{11} k_{\pm 0}^3) = \frac{\pi^2 k_0^6}{3k_0^2 x^3 + q^2(\pi^2 x - x^3/2)}, \quad (\text{A25})$$

$$(\phi_{\pm}^{01} k_{\pm 0}^3) = \frac{\pi^2 k_0^6}{2} \frac{2k_0^2 x^3 + q^2(\pi^2 x - x^3/2)}{[q^2 x][3k_0^2 x^3 + q^2(\pi^2 x - x^3/2)]}. \quad (\text{A26})$$

### References

- 1) C. Zener, Phys. Rev. **81** (1951), 446; **82** (1951), 403; **83** (1951), 299; **85** (1951), 324.
- 2) T. Kasuya, Prog. Theor. Phys. **16** (1956), 45.
- 3) K. Yosida, Phys. Rev. **106** (1957), 893.
- 4) T. Kasuya, Prog. Theor. Phys. **16** (1956), 58.
- 5) K. Yosida, Phys. Rev. **107** (1957), 396.
- 6) T. Kasuya, Prog. Theor. Phys. **20** (1958), 980.
- 7) T. Kasuya and K. Yamada, Jour. Phys. Soc. Japan **14** (1959), 416, T. Kasuya, Bussei Buturi Koza (Kyoritusha) vol 7. p. 241.
- 8) For example, A. H. Wilson, *The Theory of Metals*, Cambridge, (1953).
- 9) For example, H. H. Potter, Proc. Phys. Soc. **49** (1937), 671.
- 10) R. W. Schmitt and I. S. Jacobs, Canad. Jour. Phys. **34** (1956), 1285.
- 11) D. K. C. MacDonald and W. B. Pearson, Proc. Roy. Soc. **219** (1953), 373.  
S. Tamura, Jour. Phys. Soc. Japan **14** (1959), 541.