The theory of the magnetization process in $CeRu_2Si$

1991 J. Phys.: Condens. Matter 3 9915

(http://iopscience.iop.org/0953-8984/3/49/008)

J. Phys.: Condens. Matter 3 (1991) 9915–9928. Printed in the UK

The theory of the magnetization process in $CeRu_2Si_2$

Rikio Konno†
Institute for Solid State Physics, University of Tokyo, Tokyo, Roppongi 106, Japan and Industrial Research Institute of Kanagawa Prefecture, Kanazawa-ku, Yokohama 236, Japan

Received 23 January 1991, in final form 13 May 1991

Abstract. A metamagnetic-like transition has been reported by Haen *et al* (1987) for the magnetization curve in $CeRu_2Si_2$ with a large anisotropy. We introduce here a weakly interacting Kondo model, including crystalline field splitting, which is composed of a collection of SU(N_c) Kondo models and a RKKY interaction between the localized spins of the Ce ions. We investigate to what extent the metamagnetic-like behaviour can appear on the basis of this simplified model. The model has the advantage that we can derive the effective single site SU(N_c) Kondo Hamiltonian with the use of the mean field approximation for the intersite coupling. Then, using the exact solution of the effective Hamiltonian, we investigate the magnetization process of the ground state. With the set of appropriate crystalline field parameters determined from the analysis of the specific heat, we have obtained the metamagnetic-like transition at a reasonable magnetic field with the large anisotropy qualitatively in agreement with the experimental data of $CeRu_2Si_2$. However, to achieve this it was necessary to use a large Kondo temperature and a ferromagnetic RKKY interaction. These discrepancies may be traced back to the mean field approximation for the intersite correlations.

1. Introduction

A certain class of materials in both Ce compounds and U compounds has been investigated intensively. One of the features of these materials is that the effective mass determined by the linear coefficient of the specific heat is greater than the bare electron mass: it ranges from several hundred to one thousand times larger than for normal metals. Therefore, we call these materials heavy fermion (heavy electron) systems (Lee *et al* 1986, Fulde *et al* 1988). Correspondingly, the paramagnetic susceptibility of these materials is also larger than for normal metals.

The electronic resistivity in the Ce compounds belonging to this class of materials shows a Kondo-like effect at high temperatures. The Kondo effect is a phenomenon in which the resistivity is increased as ln T with decreasing temperatures by the exchange interaction between 4f-electrons and conduction electrons (Kondo 1964). In addition, the susceptibility obeys the Curie–Weiss law at high temperatures. On the other hand, at low temperatures the electronic resistivity is proportional to T^2, and the susceptibility shows Pauli paramagnetism at low temperatures: a typical Fermi liquid behaviour.

† Present address: Institute of Physics, University of Tsukuba, Tsukuba, Ibaraki 305, Japan.

0953-8984/91/499915 + 14 $03.50

The magnitude of the Pauli paramagnetic susceptibility is large, corresponding to the previously mentioned large linear coefficient of specific heat at low temperatures.

We can assume that the 4f-electrons in these materials are almost localized at Ce-sites because of the strong electron correlation. Then, the degree of freedom of the 4f-electrons can be represented by localized spins at the Ce-sites. The low-temperature behaviour of the single-site Kondo system is characterized by the formation of a singlet ground state where the localized spin is quenched by the conduction electron's spin polarization through antiferromagnetic exchange interaction. The characteristic temperature is called the Kondo temperature T_K, which represents the binding energy of the singlet (Yosida 1966). The 4f-electrons have orbital degeneracy and there is a strong crystalline field in the Ce compounds. The Kondo temperature for an orbitally degenerate case is generally high, while the crystalline field suppresses it (Yamada *et al* 1984, Hanzawa *et al* 1985).

On the other hand, in a lattice, the indirect exchange interaction mediated by conduction electrons, the Ruderman–Kittel–Kasuya–Yosida (RKKY) interaction, acts between 4f-electron spins at Ce-sites (Ruderman and Kittel 1954, Kasuya 1956, Yosida 1957). There exists a competition between the single site Kondo effect and the RKKY interaction in the Ce compounds. Various interesting physical phenomena appear as a result of this competition.

Recently, it was reported that in $CeRu_2Si_2$ a metamagnetic-like transition appears with the magnetic field $H^* = 80$ kOe at low temperatures from 1.75 K to 10 K. An interesting aspect of this transition is that it occurs even though there is no magnetic ordering in $CeRu_2Si_2$. The metamagnetic behaviour is observed when the magnetic field is applied parallel to the c axis (Haen *et al* 1987). However, there is no metamagnetic anomaly with the magnetic field applied perpendicular to the c axis. Furthermore, we have a large anisotropy of the magnetization in $CeRu_2Si_2$. The anomalies in the magnetoresistivity and the Hall-resistivity are also observed at about $H^* = 80$ kOe with the magnetic field parallel to the c axis but, again, with no anomaly in the field perpendicular to the c axis. The temperature dependence of the longitudinal susceptibility obeys the Curie–Weiss law at high temperatures. However, it deviates from the Curie–Weiss law at low temperatures and then shows a maximum at about 10 K. This anomaly is interpreted as an appearance of an antiferromagnetic correlation and appears in the low-temperature regime.

The specific heat of $CeRu_2Si_2$ shows two maxima. The anomaly at low temperatures is due to the Kondo effect and the one at high temperatures is due to crystalline field splitting. From specific heat data, it is estimated that the Kondo temperature for the ground doublet is $T_K = 24.4$ K and that the energy splitting between the first excited state and the ground doublet is about 220 K (Besnus *et al* 1985).

The purpose of this paper is an attempt to make a detailed analysis of the mechanism of the metamagnetic-like behaviour of $CeRu_2Si_2$. The structure of this paper is the following. We explain a model in section 2 which is used to describe the metamagnetic-like transition of $CeRu_2Si_2$. For the model, the Bethe ansatz method (Bethe 1931, Yang 1967, Andrei *et al* 1983, Tsvelick and Wiegman 1983, Hewson and Rasul 1983, Schlottman 1984, Kawakami and Okiji 1985, Okiji and Kawakami 1986, Okiji 1987) can be applied when we treat the RKKY interaction by a mean field approximation. In section 3, we analyse the results of the calculation for the magnetic field dependence of the magnetization using the exact solution. The model used here gives a description of the metamagnetic-like transition for $CeRu_2Si_2$. However, at the same time, the detailed analysis reveals that we need a set of parameters which is different from those naturally

obtained from the experiments. In section 4 we summarize the results and discuss the possible origin of the above mentioned discrepancy. We use the unit of the Boltzmann constant $k_B = 1$ throughout this paper.

2. Model

2.1. Model

The crystal structure of Ce in $CeRu_2Si_2$ is a body-centred tetragonal lattice, and assume that there is one 4f-electron per Ce ion in $CeRu_2Si_2$. Then, the multiplet of $CeRu_2Si_2$ is split into the state of total angular momentum $j = 5/2$ and the state of $j = 7/2$. Only the lowest ground multiplet of $j = 5/2$ is taken into account throughout this paper because the energy levels of $j = 7/2$ are much higher than the ones of $j = 5/2$ (about 0.3 eV from the measurement of XPS) (Lee *et al* 1986, Fulde *et al* 1988). The degeneracy of the ground multiplet of $j = 5/2$ is partly removed by the existence of a strong crystalline field, as will be mentioned in section 2.2.

The Anderson lattice model is frequently used to describe heavy fermion properties in Ce compounds (Anderson 1961). When there is a localized 4f-electron in a Ce-site, with the strong electron correlation as mentioned in the introduction, the energy level of the 4f-electron is considered to be much lower than the Fermi energy, and the Anderson lattice model then reduces to a Kondo lattice model. In the lattice, the spin polarizations of the conduction electrons (Kondo clouds) of the Kondo singlets at various sites interfere. It is very important to consider the overlap integral of the Kondo clouds but it is known to be difficult. In this paper, we adopt an approach from the dilute limit for Ce ions instead of using the real Kondo lattice model. It means that the Kondo clouds around different Ce-sites are assumed to be independent of each other. Therefore, we consider a collection of SU(N_c) Kondo models (Coqblin and Schrieffer 1969) where the crystalline field and the orbital degeneracy ($N_c = 2j + 1 = 6$) are taken into account. On the other hand, to include an effect of intersite coupling we consider that the RKKY interaction acts between the localized spins of Ce. Once the RKKY interaction is included, the model is not exactly solvable anymore. However, when we treat the RKKY interaction with a mean field approximation, we can apply the exact solution of the Bethe ansatz to this weakly interacting SU(N_c) Kondo model.

Cox has studied a similar model. However, he has not included the crystalline field which is essential and important in considering the metamagnetic-like behaviour of $CeRu_2Si_2$, and he used the $1/N_c$ expansion instead of the exact solution (Cox 1987).

The Hamiltonian of this weakly interacting SU(N_c) Kondo model can be written as follows:

$$H = \sum_i H^{(i)}_{\text{Kondo}} - \sum_{\langle i,j \rangle} K_{i,j} \boldsymbol{S}_i \cdot \boldsymbol{S}_j. \tag{2.1}$$

$H^{(i)}_{\text{Kondo}}$ is the SU(N_c) Kondo Hamiltonian for the ith Ce-site. $\boldsymbol{S}_i$ are spin operators of f-electrons and K_{ij} are the RKKY interactions among them. The explicit form of $H^{(i)}_{\text{Kondo}}$ is

$$H^{(i)}_{\text{Kondo}} = \sum_{k,m} \varepsilon_k c^\dagger_{km} c_{km} + 2J \sum_{k,m} \sum_{k',m'} c^\dagger_{k'm'} c_{km} a^\dagger_m a_{m'} - g\mu_B H_0 \sum_m m a^\dagger_m a_m + H_{\text{cry}}(J > 0) \tag{2.2}$$

where ε_k is a kinetic energy for the conduction electrons, J is an exchange interaction between the conduction electrons and a localized f-spin, H_0 is a magnetic field, g is the Lande-factor ($g = 6/7$ for Ce^{3+}), and μ_B is the Bohr magneton. $c^\dagger_{km}$ and c_{km} are, respectively, a creation operator and an annihilation operator of a conduction electron with a wavevector k and with a magnetic quantum number m. $a^\dagger_m$ and a_m are, respectively, a creation operator of a 4f-electron and an annihilation operator of a 4f-electron. H_{cry} is a crystalline field Hamiltonian which we will discuss in detail in section 2.2. $H^{(i)}_{Kondo}$ without H_{cry} is invariant under the transformation $c_{km} \to U_{mm'}c_{km'}$, $a_m \to U_{mm'}a_{m'}$ where U is an N_c-dimensional unitary matrix. The symmetry of $H^{(i)}_{Kondo}$ is $SU(N_c)$. Therefore $H^{(i)}_{Kondo}$ is called the $SU(N_c)$ Kondo model.

We may use a molecular field approximation for the RKKY interaction if we assume that the f-electron spins for Ce-sites weakly interact with each other. Then, the internal field $\boldsymbol{h}_{in}$ which is induced by the RKKY interaction is

$$\boldsymbol{h}_{in} = \frac{1}{g\mu_B}\sum_j K_{ij}\langle \boldsymbol{S}_j\rangle. \tag{2.3}$$

Now, the Hamiltonian of the ith Ce-site is

$$H^{(i)}_{MF} = \sum_{k,m}\varepsilon_k c^\dagger_{km}c_{km} + 2J\sum_{k,m}\sum_{k',m'} c^\dagger_{k'm'}c_{km}a^\dagger_m a_{m'} - g\mu_B\sum_{m,m'}\boldsymbol{h}a^\dagger_m(\boldsymbol{S})_{m,m'}a_{m'} + H_{cry} \tag{2.4}$$

$$\boldsymbol{h} = \boldsymbol{H}_0 + \boldsymbol{h}_{in} \tag{2.5}$$

$$\boldsymbol{H}_0 = \boldsymbol{h} - \frac{1}{g\mu_B}\sum_j K_{ij}\langle \boldsymbol{S}_j\rangle \tag{2.6}$$

where $\boldsymbol{h}$ is the effective field including both the external field and the internal field. When a uniform magnetic field is applied to $CeRu_2Si_2$, $\langle \boldsymbol{S}_j\rangle$ does not depend on j. On the other hand, the RKKY interaction constant K_{ij} depends only on $i - j$. Therefore, the internal field $\boldsymbol{h}_{in}$ is independent of the Ce-sites. Equation (2.6) can be rewritten as follows:

$$\boldsymbol{H}_0 = \boldsymbol{h} - (1/g\mu_B)K(0)\boldsymbol{M} \tag{2.7}$$

where $K(0)$ is $\Sigma_j K_{ij}$, and $\boldsymbol{M}$ is the magnetization per Ce ion. Equation (2.4) can be solved by the Bethe ansatz method. We can calculate the magnetization with the use of the exact solution in the field $\boldsymbol{h}$. Then, the magnetic field dependence of the magnetization is obtained from the relation between the magnetic field and the magnetization of (2.7).

2.2. *Hamiltonian for the crystalline field*

The crystal structure of Ce in $CeRu_2Si_2$ is a body-centred tetragonal lattice, and the Ce ions are subject to a crystalline field. We consider the Hamiltonian for the crystalline field here and will describe the Hamiltonian of the crystalline field for the ground multiplet of $j = 5/2$ in terms of the spin operators. The independent product forms of two spin operators are up to the fifth order. The Hamiltonian of the crystalline field

should be invariant under symmetry operations of the D_{4h} group. Therefore, second-order terms and fourth-order terms of the spin operators only may appear in the Hamiltonian of the crystalline field. The only second-order term in the Hamiltonian is

$$V_{T_2}[S_x^2 + S_y^2 - 2S_z^2] \tag{2.8}$$

where V_{T_2} is the coupling constant of the second-order tetragonal field. We have chosen the c axis as the z axis of the spin space. Similarly, the fourth-order terms in the Hamiltonian of the crystalline field are

$$V_c[S_x^4 + S_y^4 + S_z^4 - \tfrac{1}{5}S(S+1)(3S(S+1) - 1)] \tag{2.9a}$$

$$V_{T_4}[S_x^4 + S_y^4 - 2S_z^4] \tag{2.9b}$$

$$V_{T_4'}[S_y^2S_z^2 + S_z^2S_x^2 - 2S_x^2S_y^2] \tag{2.9c}$$

where V_{T_4} and $V_{T_4'}$ are the coupling constants of the fourth order for the tetragonal crystalline field and V_c is a coupling constant for a cubic crystalline field. Since $V_{T_4'}[S_y^2S_z^2 + S_z^2S_x^2 - 2S_x^2S_y^2]$ is expressed by a linear combination of $V_{T_2}[S_x^2 + S_y^2 - 2S_z^2]$ and $V_{T_4}[S_x^4 + S_y^4 - 2S_z^4]$, in total there are three independent terms in the Hamiltonian of the crystalline field. Therefore, the most general Hamiltonian of the crystalline field H_{cry} is

$$H_{cry} = V_{T_2}[S_x^2 + S_y^2 - 2S_z^2] + V_c[S_x^4 + S_y^4 + S_z^4 - \tfrac{1}{5}S(S+1)(3S(S+1) - 1)] + V_{T_4}[S_x^4 + S_y^4 - 2S_z^4]. \tag{2.10}$$

We can diagonalize

$$H_{cry} - g\mu_B h \sum_{m=-j}^{j} m \tag{2.11}$$

whose eigenvalues ω are as follows:

$$\omega = \begin{cases} -(a+b) - \frac{1}{2}\tilde{h} \\ -(a+b) + \frac{1}{2}\tilde{h} \\ (a+b+\tilde{h})/2 + [((a-b)/2) + 2\tilde{h})^2 + c^2]^{1/2} \\ (a+b+\tilde{h})/2 - [((a-b)/2) + 2\tilde{h})^2 + c^2]^{1/2} \\ (a+b-\tilde{h})/2 + [((a-b)/2) - 2\tilde{h})^2 + c^2]^{1/2} \\ (a+b-\tilde{h})/2 - [((a-b)/2) - 2\tilde{h})^2 + c^2]^{1/2}. \end{cases} \tag{2.12}$$

In the above expression, a, b, c are given by

$$a = -10V_{T_2} + 3V_c - 70V_{T_4} \tag{2.13a}$$

$$b = 2V_{T_2} - 9V_c + 20V_{T_4} \tag{2.13b}$$

$$c = 3[5(V_c + V_{T_4})]^{1/2} \tag{2.13c}$$

and $\tilde{h}$ is given by

$$\tilde{h} = g\mu_B h. \tag{2.13d}$$

When $h = 0$, the six states are grouped into three Kramers doublets.

$$\omega_0 = -(a + b) \tag{2.14a}$$

$$\omega_+ = (a + b)/2 + ([(a - b)/2]^2 + c^2)^{1/2} \tag{2.14b}$$

$$\omega_- = (a + b)/2 - ([(a - b)/2]^2 + c^2)^{1/2}. \tag{2.14c}$$

Wave-functions of the doublets are

$$|0\rangle = |\pm\tfrac{1}{2}\rangle \tag{2.15a}$$

$$|+\rangle = \sin\theta|\pm\tfrac{5}{2}\rangle - \cos\theta|\mp\tfrac{3}{2}\rangle \tag{2.15b}$$

$$|-\rangle = \cos\theta|\pm\tfrac{5}{2}\rangle + \sin\theta|\mp\tfrac{3}{2}\rangle \tag{2.15c}$$

where the mixing angle θ is defined by

$$\cos\theta = (\omega_- - b)/[(b - \omega_-)^2 + c^2]^{1/2} \tag{2.16a}$$

$$\sin\theta = c/[(b - \omega_-)^2 + c^2]^{1/2}. \tag{2.16b}$$

Now, we consider a magnetic field parallel to the x axis. Then, the Hamiltonian is of the form

$$H_{cry} - g\mu_B H S_x = \begin{pmatrix} a & -(\sqrt{5}/2)\tilde{h} & 0 & 0 & c & 0 \\ -(\sqrt{5}/2)\tilde{h} & b & -\sqrt{2}\tilde{h} & 0 & 0 & c \\ 0 & -\sqrt{2}\tilde{h} & -(a+b) & -\tfrac{3}{2}\tilde{h} & 0 & 0 \\ 0 & 0 & -\tfrac{3}{2}\tilde{h} & -(a+b) & -\sqrt{2}\tilde{h} & 0 \\ c & 0 & 0 & -\sqrt{2}\tilde{h} & b & -(\sqrt{5}/2)\tilde{h} \\ 0 & c & 0 & 0 & -(\sqrt{5}/2)\tilde{h} & a \end{pmatrix}. \tag{2.17}$$

The diagonalization in this case has been done numerically.

By treating the crystalline fields as an external field, we can use the Bethe ansatz solution of the SU(N_c) Kondo model for the problem with the crystalline field in (2.4). We can calculate the magnetization using the exact solution for the ground state by following Hewson *et al* (1983) and Kawakami and Okiji (1985). In the next section, we will mention and discuss the results of this calculation.

3. Results

In the present approach, the model of weakly interacting Kondo centres is treated by a mean field approximation for the intersite RKKY interaction as mentioned in section 2.1. Therefore, after discussing the magnetic field dependence of magnetization for the

effective single impurity problem using the exact solution, we can take account of the RKKY interaction. Here we investigate how far the metamagnetic-like behaviour can occur on the basis of this simplified model. Hence, the results are compared with the experiments.

3.1. General discussions

First, we investigate the magnetization curve in the presence of the crystalline field. It should be noted that we have kept $CeRu_2Si_2$ in mind throughout the following discussion. Cerium ions in $CeRu_2Si_2$ form a body-centred tetragonal lattice. The energy levels in the Ce ions are split into three doublets by the crystalline field as mentioned in section 2.2. We then have the following three cases depending on the magnitude of V_{Γ_2}, V_{Γ_4}, V_c: (1) $\omega_- \leqslant \omega_0 \leqslant \omega_+$, (2) $\omega_- \leqslant \omega_+ \leqslant \omega_0$, (3) $\omega_0 \leqslant \omega_- \leqslant \omega_+$. Experimentally, it has been found that in $CeRu_2Si_2$ the magnetization in the magnetic field along the c axis is larger than that in the field perpendicular to the c axis (Haen *et al* 1987). Therefore, we will only discuss the magnetization curve in cases (1) and (2), because in case (3) the plane perpendicular to the c axis is an easy plane of magnetization and in contradiction to the experiment.

3.1.1. $\omega_- \leqslant \omega_0 \leqslant \omega_+$. In order to investigate the magnetization curve, we have to determine the crystalline field parameters, which were introduced in section 2.2. The crystalline field parameters are related to the energy level scheme. The energy level scheme of the three doublets split by the crystalline field can, in principle, be determined by the neutron scattering experiment. However, at present there are no neutron scattering experiments available with which the energy level scheme of the three doublets can be determined. Therefore, we will estimate the crystalline field parameters from the magnitude of the crystalline field splitting $\Delta_{10} = 220$ K and $\Delta_{20} = 1000$ K (Besnus *et al* 1985). Δ_{10} is the energy splitting between the first excited state and the ground state, and Δ_{20} is between the second excited state and the ground state. As we have discussed in section 2.2, there are three parameters for the crystalline field in this case; one cubic field term and the two tetragonal field terms. From (2.14a, b, c), Δ_{10}, Δ_{20} are related to the parameters of the crystalline field a, b, c:

$$b = -\tfrac{2}{3}\Delta_{10} + \tfrac{1}{3}\Delta_{20} - a \tag{3.1a}$$

$$c = \pm[(\tfrac{1}{2}\Delta_{20})^2 - (a + \tfrac{1}{3}\Delta_{10} - \tfrac{1}{6}\Delta_{20})^2]^{1/2}. \tag{3.1b}$$

As the argument of a square root must be positive, the range of a is determined as follows:

$$-\tfrac{1}{3}\Delta_{10} - \tfrac{1}{3}\Delta_{20} \leqslant a \leqslant -\tfrac{1}{3}\Delta_{10} + \tfrac{2}{3}\Delta_{20}. \tag{3.1c}$$

We cannot determine the three parameters of the crystalline field from the two data of $\Delta_{10} = 220$ K and $\Delta_{20} = 1000$ K. However, as a working hypothesis we take only the lowest (second)-order tetragonal term for the crystalline field and the cubic term. Then, V_{Γ_2} and V_c can be determined uniquely. The condition that the c axis is the easy axis of magnetization is $V_{\Gamma_2} > 0$. In this way, we can obtain the following values: $V_{\Gamma_2} = 13.5$ K, $V_c = -49.1$ K, and $V_{\Gamma_4} = 0$ K.

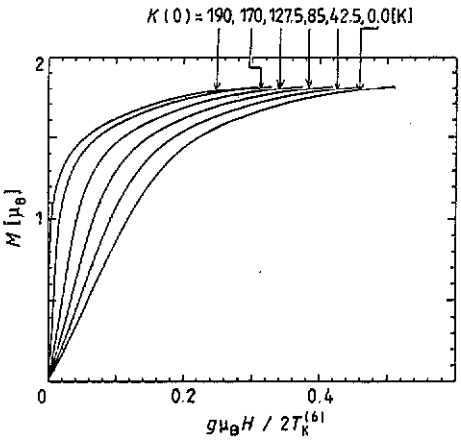

Figure 1. The magnetization curves with the magnetic field applied parallel to the c axis. The crystalline field parameters used are $V_{T_2} = 13.5$ K, $V_c = -49.1$ K, $V_{T_4} = 0.0$ K. The Kondo temperature for the sixfold degeneracy is $T_K^{(6)} = 850$ K. The RKKY interaction constants $K(0) = 0.0$ K, 42.5 K, 85 K, 127.5 K, 170 K, 190 K are used for the magnetization curves from right to left.

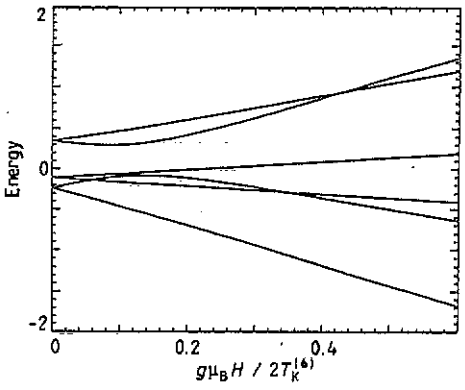

Figure 2. The magnetic field dependence of each energy level with the magnetic field applied parallel to the c axis for the same coupling constants as in figure 1, $V_{T_2} = 13.5$ K, $V_c = -49.1$ K, $V_{T_4} = 0.0$ K, $T_K^{(6)} = 850$ K. The RKKY interaction constant is $K(0) = 0.0$ K. We normalize the scale of the vertical axis by $2T_K^{(6)} = 1700$ K.

We need the Kondo temperature $T_K^{(6)}$ for the sixfold degenerate energy levels to be the energy scale of the SU(N_c) Kondo model (Hanzawa *et al* 1985), where

$$T_K^{(6)} = D \exp(-1/N_c \rho_0 J) \tag{3.2}$$

and where ρ_0 is a density of states, and D is the bandwidth of the conduction band. Hanzawa *et al* (1985) derived the relation between $T_K^{(6)}$ and the effective Kondo temperature $T_K^{(2)}$ for the ground doublet in the presence of the crystalline field,

$$T_K^{(6)} = (\Delta_{10}\Delta_{20}T_K^{(2)})^{1/3} \tag{3.3}$$

for the SU(N_c) Kondo lattice model. The derivation is based on the Yoshida theory and the poor man's scaling theory. Lacerda *et al* (1989) estimated $T_K^{(6)} \simeq 170$ K using $\Delta_{10} = 220$ K, $\Delta_{20} = 1000$ K and $T_K^{(2)} = 24.4$ K in (3.3).

Figure 1 shows the magnetization curve with the magnetic field applied parallel to the c axis for $V_{T_2} = 13.5$ K, $V_c = -49.1$ K, $V_{T_4} = 0$ K, $T_K^{(6)} = 850$ K where the coupling constant of the RKKY interaction is varied as $K(0) = 0.0$, 42.5, 85.0, 127.5, 170, 190 K. Since the intersite RKKY interaction is treated in the mean field approximation, it is necessary to study the case $K(0) = 0$ K in detail in order to understand the behaviour of the magnetization. The magnetization curve non-linearly increases with an increase of the magnetic field from $g\mu_B H/(2T_K^{(6)}) = 0.0$ to $g\mu_B H/(2T_K^{(6)}) = 0.06$. There is an inflection point at $g\mu_B H/(2T_K^{(6)}) \simeq 0.06$ similar to the case where there is an absence of both the crystalline field and the RKKY interaction (Hewson and Rasul 1983).

To understand the origin of the non-linear increase of the magnetization, figure 2 shows the energy levels split by the magnetic field applied parallel to the c axis when $K(0) = 0$ K. The ground state doublet is the linear combination of $|\frac{5}{2}\rangle$ and $|-\frac{5}{2}\rangle$. For the set of parameters, the main component of the ground state is $|\frac{5}{2}\rangle$ at zero field and its

weight increases as the magnetic field is increased. The energy of the ground state decreases faster than linearly in the magnetic field. Correspondingly, we find that the number of electrons in the ground state becomes much larger than those in the other states. These two factors together cause a non-linear increase of the magnetization curve similar to that when there is an absence of the crystalline field.

However, this upturn of the magnetization curve is less remarkable than in the case when there is an absence of the crystalline field. This upturn becomes sharper when the population in the level other than the ground doublet is larger, at $H = 0$. Under the condition that there is a strong anisotropy this is possible only when the magnitude of $2T_K^{(6)}$ is comparable to the crystalline field splitting.

Now we introduce the RKKY interaction. As seen from figure 1, the increase in magnetization becomes steeper as $K(0)$ is increased. Note here that the scale of the magnetic field is very large. For sufficiently large $K(0)$, we obtain the metamagnetic-like behaviour and the eventually real metamagnetic transition is realized. The figure with an enlarged scale in the relevant field range will be shown later to compare the experimental results.

We have also studied the case with the field perpendicular to the c axis. In this case, as is expected, the magnetization grows much slower than the case when the field is parallel to the c axis. In this case too the ground level of the multiplet decreases continuously with increasing field. Namely, as the field is increased the wave-function of the ground level changes continuously from the form given by (2.15c) to $(1/\sqrt{2})(|\frac{1}{2}\rangle + |-\frac{1}{2}\rangle)(H \| x)$. We may think that a kind of spin–flop process takes place in the anisotropy field in this case. Moreover, this spin–flop takes place continuously. The usual spin–flop process happens suddenly in the spin system where there is long range order.

3.1.2. $\omega_- \leqslant \omega_+ \leqslant \omega_0$. In this situation a peculiar magnetization curve is realized when the level crossing occurs in the field. This is possible when $|\pm\frac{3}{2}\rangle$ has a larger weight than $|\mp\frac{5}{2}\rangle$ at zero field. As an example we show the magnetization curve in figure 3 for the parameters $V_{T_2} = 85$ K, $V_c = 20$ K, $V_{T_4} = -10$ K. We have chosen the parameters so that they are consistent with the experimental observations as well as the above mentioned condition. The level scheme determined by these crystalline field parameters is $\Delta_{10} = 180$ K and $\Delta_{20} = 540$ K. The level crossing in the ground doublet is seen in figure 4 where the level scheme in the field is plotted. The magnetization increases in two steps. The second sharp increase occurs near the field strength of the level crossing. A similar step-wise increase of the magnetization is reported by Kawakami *et al* (1989) for the hexagonal system with the equidistance level scheme of $|\pm\frac{1}{2}\rangle, |\pm\frac{3}{2}\rangle, |\pm\frac{5}{2}\rangle$. When the RKKY interaction is increased, the two steps become sharper, and for this set of parameters, the first step is sharper than the second. This eventually leads to a ferromagnetic state before showing the real metamagnetic transition.

3.2. Comparison of the theory with experiments

Haen *et al* (1987) reported for $CeRu_2Si_2$ that the magnetization increases non-linearly with an increase of the magnetic field when it is applied parallel to the c axis. In addition, they reported that the metamagnetic-like transition appears at $H^* \simeq 80$ kOe. On the other hand, there is no metamagnetic anomaly with the magnetic field applied perpendicular to the c axis (Haen *et al* 1987). These behaviours are not consistent with the

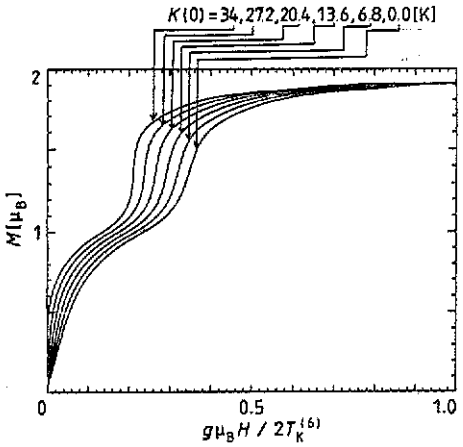

Figure 3. The magnetization curves with the magnetic field applied parallel to the c axis. The crystalline field parameters used are $V_{T_2} = 85$ K, $V_c = 20$ K, $V_{T_4} = -10$ K. The Kondo temperature for the sixfold degeneracy is $T_K^{(6)} = 170$ K. The magnetization curves are for the RKKY interaction constants $K(0) = 0.0$ K, 6.8 K, 13.6 K, 20.4 K, 27.2 K, 34 K from right to left.

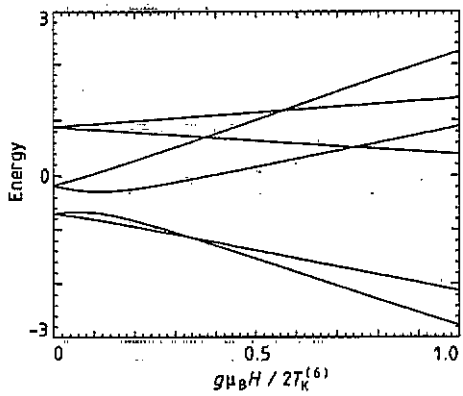

Figure 4. The magnetic field dependence of each energy level with the magnetic field applied parallel to the c axis for $V_{T_2} = 85$ K, $V_c = 20$ K, $V_{T_4} = -10$ K, $T_K^{(6)} = 170$ K and $K(0) = 0.0$ K. We normalize the scale of the vertical axis by $2T_K^{(6)} = 340$ K.

two-step magnetization process. Therefore, case 1 in the preceding section is considered to be a candidate to explain the experimental results.

A comparison between the theory and the experiments is shown in figure 5. The full curves are the calculated curves with the crystalline field parameters $V_{T_2} = 13.5$ K, $V_c = -49.1$ K, $V_{T_4} = 0$ K, the Kondo temperature for the sixfold degeneracy $T_K^{(6)} = 850$ K, and the RKKY interaction constant $K(0) = 190$ K used in the example for case 1. The open circles are the experimental data from Haen *et al* (1987). For the magnetic field applied parallel to the c axis the theoretical magnetization curve in figure 5 increases non-linearly and shows the metamagnetic-like transition at $H^* \simeq 70$ kOe On the other hand, the magnetization curve is linear for the magnetic field perpendicular to the c axis. There is no anomaly of the magnetization in this direction. These features are qualitatively consistent with the experiments.

For the set of crystalline field parameters, we used the value obtained from the reported values of $\Delta_{10} = 220$ K and $\Delta_{20} = 1000$ K by the procedure described in section 3.1. However, it seems there is an ambiguity in determining Δ_{20} and the value of 1000 K seems to be larger than the crystalline field splitting of typical Ce compounds. Therefore, as a second set of parameters, we assume that $\Delta_{20} = 2\Delta_{10} = 440$ K with $\Delta_{10} = 220$ K fixed. Then, we obtain $V_{T_2} = 13.24$ K, $V_c = -17.66$ K, $V_{T_4} = 0.0$ K by the same procedure. For this set of parameters we need $2T_K^{(6)} = 1330$ K and $K(0) = 153$ K to obtain the metamagnetic-like transition. The results for the second set of parameters are shown in figure 6 with qualitatively similar results to figure 5.

The RKKY interaction constant $K(0)$ and the Kondo temperature for the sixfold degeneracy $T_K^{(6)}$ are not independent. In the second-order perturbation theory the RKKY interaction is expressed by

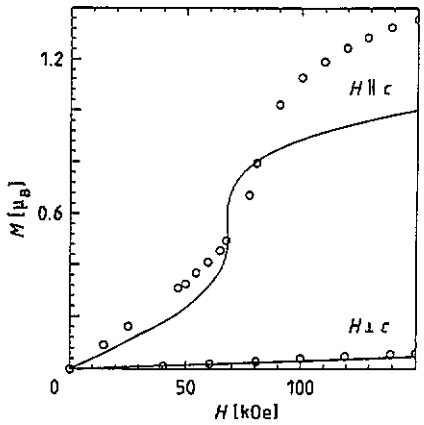

Figure 5. Magnetization versus the magnetic field. $H \parallel c (H \perp c)$ represents the magnetization curve in the magnetic field applied parallel (perpendicular) to the c axis. The crystalline field parameters used are $V_{\Gamma_2} = 13.5$ K, $V_c = -49.1$ K, $V_{\Gamma_4} = 0.0$ K. The Kondo temperature for the sixfold degeneracy is $T_K^{(6)} = 850$ K. The RKKY interaction constant is $K(0) = 190$ K. The experimental data by Haen *et al* (1987) are shown by the open circles.

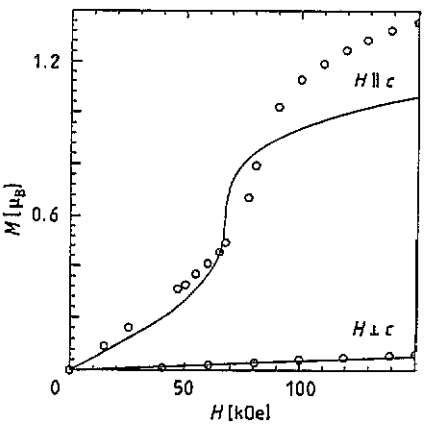

Figure 6. Magnetization versus the magnetic field. $H \parallel c (H \perp c)$ represents the magnetization curve in the magnetic field applied parallel (perpendicular) to the c axis. The crystalline field parameters used are $V_{\Gamma_2} = 13.24$ K, $V_c = -17.66$ K, $V_{\Gamma_4} = 0.0$ K. The Kondo temperature for the sixfold degeneracy is $T_K^{(6)} = 665$ K. The RKKY interaction constant is $K(0) = 153$ K. The experimental data by Haen *et al* (1987) are shown by the open circles.

$$-\tfrac{1}{2}\rho_0 J^2 \sum_{i,j} \left[\sum_Q f(Q)\, e^{iQ\cdot R_{ij}} \right] S_i \cdot S_j \tag{3.4}$$

where R_{ij} is the difference between the position of the ith and jth Ce-site and $f(Q)$ is the normalized susceptibility of the conduction band. In the electron gas model, $f(Q)$ is given by

$$f(Q) = 1 + [(1 - x^2)/2x] \ln|(1 + x)/(1 - x)|$$

where

$$x = Q/2k_F$$

with k_F being a Fermi wave-number. The Fourier transform of the $K(Q)$ is

$$K(Q) = \tfrac{1}{2}\rho_0 J^2 [f(Q) - \bar{f}] \tag{3.5}$$

where we subtract the average of $f(Q)$ to exclude the term $i = j$ from the summation in (3.4). As mentioned in section 2.1, we only take account of the component $Q = 0$. The magnitude of the uniform component of the RKKY interaction is of the order of $D(\rho_0 J)^2$ as is seen from (3.5). The uniform component $K(0)$ is given by $T_K^{(6)}$ through

$$K(0) = D/(N_c \log(D/T_K^{(6)}))^2.$$

When we use the representative values $D = 10000$ K, $K(0) = 46$ K for the first example, $T_K^{(6)} = 850$ K, and $K(0) = 38$ K for the second example $T_K^{(6)} = 665$ K. The magnitude of the RKKY interaction is about a quarter of $K(0)$ which is used to reproduce the metamagnetic-like behaviour. This suggests that we had to use a larger magnitude of the RKKY interaction than that naturally reduced from $T_K^{(6)}$ in the present model.

Here we summarize the above result. If we use the set of parameters for the crystalline field, the metamagnetic-like behaviours obtained with the present model are qualitatively in agreement with experiments. In particular, the value of H^* and anisotropy are reasonable. Therefore, we may say that the metamagnetic-like behaviour itself can be obtained from the simple weakly interacting Kondo model with a crystalline field splitting (an approach from a low concentration limit of Ce ions). However, at the same time we should mention that we had to use a larger $T_K^{(6)}$ as compared with experiments and a large positive (ferromagnetic) $K(0)$. This means that we need an improvement of the present model and/or the approximation scheme in order to understand completely the mechanism of metamagnetic-like behaviours.

The reason that the large $T_K^{(6)}$ and $K(0)$ are required is the following. One characteristic of the metamagnetic-like transition of $CeRu_2Si_2$ is a large anisotropy of magnetization. This means that there exists the strong crystalline field in $CeRu_2Si_2$. Then, the Kondo temperature for the sixfold degeneracy $T_K^{(6)}$ is much higher than the Kondo temperature for the ground doublet $T_K^{(2)}$ which can be measured from experiments. However, $T_K^{(6)}$ is much smaller than Δ_{20} when $T_K^{(2)} \ll \Delta_{10}, \Delta_{20}$, see (3.3). To show the large anisotropy the main component of the ground doublet should be $|\frac{5}{2}\rangle$. On the other hand, in order to have metamagnetic-like transition states with smaller magnetic quantum numbers, m should be populated at $H = 0$, as we have seen in section 3.1. The population decreases rapidly with an increase of the magnetic field. This is the reason why we need a large $T_K^{(6)}$ in order to obtain the metamagnetic-like transition in the present model. However, the change of magnetization for the magnetic field is very slow for the large $T_K^{(6)}$. Therefore, we also need a large RKKY interaction constant.

In the present approach, the orbital degeneracy and crystalline field are included. The single site Kondo problem concerning these degrees of freedom is treated exactly by the Bethe ansatz method. Therefore, the discrepancy should be traced back to the intersite correlation between magnetic ions. In the present model, each of the localized spins in Ce-sites forms the singlet ground state independently. Actually, in the case of a real lattice, it is gradually becoming clear that it is essential to take account of the antiferromagnetic correlation between the localized spins. This means that the RKKY interaction has the maximum not only at $Q = 0$ but also at large Q corresponding to the antiferromagnetic wave-vector. The latter causes the quenching of the spin moment due to the quantum fluctuations when the magnetic field is small. In the present approach, we have not taken account of this effect since we used the mean field approximation for the RKKY interaction. If the antiferromagnetic correlation between Ce ions develops, the reduction of spin due to the Kondo effect can be small. In addition, it has been pointed out that phonon effects are important at the metamagnetic-like transition in $CeRu_2Si_2$ (Ohkawa 1989, Bruls *et al* 1990). So far, we have neglected these phonon effects. Therefore, we expect that we can better understand the mechanism of the metamagnetic-like behaviour in $CeRu_2Si_2$ with more realistic $T_K^{(6)}$ and $K(Q)$ if we accurately treat the antiferromagnetic correlations between Ce ions and the phonon effects.

4. Conclusion

In the present article, we have taken an approach from a low concentration limit of Ce ions in order to understand the metamagnetic-like transition observed in $CeRu_2Si_2$. The singlet ground states are assumed to be independently formed around Ce-sites. We neglect the overlap between the polarization clouds of the conduction electrons which form singlets together with the localized spins. Instead of including this interference

effect we assumed that RKKY interactions act on the localized spins of Ce. We have derived the effective Hamiltonian of a single impurity including the crystalline field and the orbital degeneracy by treating the RKKY interaction in the mean field approximation. Applying the Bethe ansatz method to this Hamiltonian, we have discussed the magnetization process for $CeRu_2Si_2$ on the basis of this Hamiltonian.

We have investigated the magnetization curve in the presence of the crystalline field and the RKKY interaction keeping in mind $CeRu_2Si_2$. The crystalline field parameters are determined by using the energy levels structure obtained from the analysis of the experimental results of specific heat. With the appropriate Kondo temperature for the sixfold degeneracy and RKKY interaction constant the metamagnetic-like behaviour is reproduced with the set of the crystalline field parameters. The metamagnetic-like behaviour appears in the case where the state $|\pm\frac{1}{2}\rangle$ is a first excited state in the energy level scheme. The metamagnetic-like behaviour obtained is qualitatively in agreement with experiments. In particular, the characteristic magnetic field H^* and the anisotropy of magnetization are reasonable. Therefore, we may say that we can explain the metamagnetic-like transition itself in the present model. At the same time, the limitation of the present model has also become clear. We needed a large ferromagnetic RKKY interaction and a large Kondo temperature for the sixfold degeneracy in order to obtain the metamagnetic-like transition. Note here that we have used the mean field approximation for the RKKY interaction, while single site effects are fully taken into account by the exact solution. In the mean field approximation, the $Q = 0$ component of the RKKY interaction is used. If we take account of the intersite correlation between Ce ions, which is neglected in the present approach, we expect that the localized spins partly compensate with each other by the antiferromagnetic (large Q) components of the RKKY interaction, and this may lead to a more reasonable value for the Kondo temperature. We have neglected the phonon effects which are important at the metamagnetic-like transition in $CeRu_2Si_2$ as mentioned in section 3.2 (Ohkawa 1989, Bruls *et al* 1990). Consequently, we need to develop a theory which takes account of the intersite correlation and phonon effects in order to obtain a better understanding of the metamagnetic-like transition in $CeRu_2Si_2$.

Acknowledgments

I am grateful to Toru Moriya and Kazuo Ueda for many helpful discussions. I would also like to thank Yoshinori Takahashi, Hirokazu Tsunetsugu, Norio Kawakami, Katsuro Hanzawa, Takashi Yanagisawa, Kiyohide Nomura and Toru Sakai for many discussions.

References

Anderson P W 1961 *Phys. Rev.* **124** 41
Andrei N, Furuya K and Lowenstein J H 1983 *Rev. Mod. Phys.* **55** 331
Besnus M J, Kappler J P, Lehmann P and Meyer A 1985 *Solid State Commun.* **55** 779
Bethe H A 1931 *Z. Phys.* **71** 205
Bruls G, Weber D, Lüthi B, Flouquet J and Lejay P 1990 *Phys. Rev.* B **42** 4329
Coqblin B and Schrieffer J R 1969 *Phys. Rev.* **185** 847
Cox D L 1987 *Phys. Rev.* B **35** 4561
Fulde P, Keller J and Zwicknagl G 1988 *Solid State Physics* vol 41 (New York: Academic) p 1
Haen P, Flouquet J, Lapierre F and Lejey P 1987 *J. Low Temp. Phys.* **67** 391

Hanzawa K, Yamada K and Yosida K 1985 *J. Magn. Magn. Mater.* **47–48** 357
Hewson A C and Rasul J W 1983 *J. Phys. C: Solid State Phys.* **16** 6799
Kasuya T 1956 *Prog. Theor. Phys.* **16** 45
Kawakami N and Okiji A 1985 *J. Phys. Soc. Japan* **54** 685
Kawakami N, Usuki T and Okiji A 1989 *J. Phys. Soc. Japan* **58** 1427
Kondo J 1964 *Prog. Theor. Phys.* **32** 37
Lacerda A, de Visser A, Haen P, Lejey P and Flouquet J 1989 *Phys. Rev.* B **40** 8759
Lee P A, Rice T M, Serene J W, Sham L J and Willkins J W 1986 *Comment. Cond. Mat. Phys.* **12** 99
Ohkawa F J 1989 *Solid State Commun.* **71** 907
Okiji A 1987 *Fermi Surface Effects* ed J Kondo and A Yosimori (Berlin: Springer) p 63
Okiji A and Kawakami N 1986 *J. Magn. Magn. Mater.* **54–57** 327
Ruderman M A and Kittel C 1954 *Phys. Rev.* **96** 99
Schlottmann P 1984 *Phys. Rev.* B **30** 1454
Tsvelick A M and Wiegmann P B 1983 *Adv. Phys.* **32** 453
Yamada K, Hanzawa K and Yosida K 1984 *Prog. Theor. Phys.* **71** 450
Yang C N 1967 *Phys. Rev. Lett.* **19** 1312
Yosida K 1957 *Phys. Rev.* **106** 893
—— 1966 *Prog. Theor. Phys.* **36** 875