Between metamagnetic transition and spin-flip behavior in Ce 122 system of (Ce-Gd)Ru2Si2

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Abstract. Aiming at getting some clues to the mechanism of meta-magnetic transition and surprisingly small magnetic moment of Ce along hard axis in CeRu2Si2, the (Ce-Gd)Ru2Si2 system where Ce was substituted by Gd were studied through magnetic properties mainly in Gd-rich regions. At Gd=0, i.e. in CeRu2Si2, the magnetic moment of Ce showed a symptom of saturation in M-H curve under H=90,000 Oe at 2 K and the Ce magnetic moment at 4.2 K can be nearly identical to that at 2 K employing 1/H plot. At Gd-rich content of 0.8, Ce magnetic moment coupled parallel to that of Gd, Ce↑Gd↑ both in easy and hard axis and the extremely smallness of Ce magnetic moment in hard axis disappeared perfectly at x=0.8. Furthermore at Gd=1, GdRu2Si2, Gd magnetic moment caused 2-step like spin-flip in both easy and hard axis.

1. Introduction

Ce122 systems have been one of the most extensively studied heavy fermion materials and have revealed various properties such as superconductivity, ferromagnetism and so on depending on the electronic states of Ce [1,2]. Consequently, the Ce 122 systems have been attracting many researchers and remain to be one of the main topics at present. CeRu2Si2, which belongs to the Ce122 systems, is one of the most famous heavy fermion compounds and is known magnetically as paramagnetic [3]. On the other hand, CeRu2Ge2 where Si is replaced by Ge shows clear ferromagnetic behaviour with Curie temperature Tc of 7.9 K [2] and many substituted systems such as CeRu2(Si-Ge)2 [4] and Ce(Ru-Rh)2Si2 [5,6] have been investigated in detail and interesting results have been obtained. Furthermore these two compounds have been treated often as a good example of investigating a nature of quantum critical point (QCT) and the CeRu2Si2 have attracted many researchers [7,8] also from this point of view.

At the beginning of the study of CeRu2Si2, Haen et al. have carried out through investigations about this compound and pointed out 2 peculiar characteristics [9]. The first is the most famous “metamagnetic” transition, that is, the Ce magnetic moment increases sharply around the magnetic field of H=77,000 Oe applied in easy c axis under 4.2 K and the second is the unexpected smallness of Ce magnetic moment (< 0.1 μB) along hard axis (H⊥c-axis). Regarding the first feature of metamagnetic transition in CeRu2Si2 along c-axis, some experiments and interpretation have been carried out and suggested [10,11]. One of the representatives was carried out using de Haas van Alphen
(dHvA) effect and it was suggested that the $4f$ electron of Ce varied from itinerant to localized nature at meta-magnetic transition field $H_m$ with the increase of magnetic field [10]. On the other hand, the soft X-ray magnetic circular dichroism (XMCD) measurement seems to have suggested a little different picture [11]. At present, there have been no decisive pictures regarding the mechanism of metamagnetic transition and the electronic state of $4f$ electron in Ce before and after this transition.

In this study, we have aimed and tried to get some clues to the mechanism about metamagnetic transition and the unexpectedly small magnetic moment of Ce in CeRu$_2$Si$_2$ by using magnetic measurement for the substituted system of (Ce-Gd)Ru$_2$Si$_2$ where Gd was employed because of the simplest ($L=0$) and strongest molecular field in Rare Earth (RE) elements.

2. Experimental Procedure

Samples studied were all prepared by arch-melting the stoichiometrically weighted constituents 5 times and the obtained poly crystals were broken into pieces with a hammer. From these small pieces, some flakes with both sides glossy were picked up and checked by Laue photographs. The samples of single crystal like flakes obtained from these procedures were of the form of sheet and the magnetically easy axis of c-axis lied perpendicular to the sheets. These single-like crystals were magnetically measured. The purities of constituent elements used were 3N for Ce and Gd, 4N for Ru and 5N for Si, respectively.

Magnetic measurements were carried out for the above-mentioned samples from 2 K to 300 K under the magnetic field up to 90,000 Oe by the use of both SQUID and VSM system of Quantum Design Corporation.

3. Results and Discussion

The measurements of magnetic properties, here focusing on the $M-H$ curves at various temperatures, were performed for mainly Gd-rich contents in both directions of easy axis and hard axis. Each result and discussion was described below, respectively.

3.1. In case of $x=0$ (i.e. CeRu$_2$Si$_2$)

At the beginning of the research of CeRu$_2$Si$_2$, Haen et al. have carried out detailed measurements regarding $M-H$ curves in both directions of magnetically easy axis ($H//c$-axis) and hard axis ($H\perp c$-axis). In their paper, they have pointed out two characteristic behaviors observed in this compound; the first is the metamagnetic transition around 77,000 Oe below 4.2 K in easy axis and this transition disappeared completely around 15 K and the second is the unexpected smallness of Ce magnetic moment in hard axis even in low temperatures.

Fig.1 (a), (b) show the present measured $M-H$ curves for $x=0$ in easy axis ($H//c$-axis) and hard axis ($H\perp c$-axis), respectively. From Fig.1 (a), the behaviors in $M-H$ at 2 and 4.2 K are roughly identical to the detailed measurement for the single crystal by Sakakibara et al. [12] and the present result at 2 K shows a clearer symptom towards saturation in the magnetic field up to 90,000 Oe in comparison with their result below $H=85,000$ Oe. As described by many researchers, the present Ce magnetic moment increases linearly with the increase of magnetic field up to around $H=50,000$ Oe even in low temperatures. In this sense that the magnetic moment is nearly proportional to magnetic field $H$, Ce magnetic moment behaves like paramagnetism, however, the value of Ce magnetic moment is not small but rather large enough to be called magnetic to some degree by considering that Ni retains about 0.6 $\mu_B$ in Ni metal and 0.2 $\mu_B$ in Ni-RE (Rare Earth) compounds [13]. In Fig.1 (b), the Ce magnetic moment behavior at 3 K in hard axis is shown. The magnetic moment of Ce in low temperatures is considerably small and about 1/30 smaller than that in easy axis. The value and linear behavior in hard axis is nearly identical to the result of Haen et al. Fig.1 (c) and (d) show the $M-1/H$ plot in easy axis at 2 K and 4.2 K, respectively. This $M-1/H$ plot is not used frequently and used conveniently to assume and determine the saturation magnetization of the sample [14]. From both results of (c) and (d), Ce magnetic moment at 4.2 K can be nearly identical to that at 2 K and this result implies that the Ce $4f$ electronic state at 4.2 K is nearly identical to that at 2 K. This result is
considered to be important to microscopical measurement since the lowest temperature obtained in the experiments can be often limited to 4.2 K because of the He cryostat.

![Figure 1](image1.png)

**Figure 1.** $M$-$H$ curves for Gd=0 compound at some temperatures. (a) and (b) in easy axis. (b) includes also in hard axis at 3 K. (c) and (d) show $M$-$1/H$ plot in easy axis.

3.2. In case of $x=0.8$ (i.e. $(\text{Ce}_{0.2}\text{Gd}_{0.8})\text{Ru}_2\text{Si}_2$)

In case of Gd content of 0.8, the $M$-$H$ curves at some temperatures between 2 K and 70 K up to the magnetic field of 70,000 Oe were measured in both directions of magnetically easy axis ($H \parallel c$ axis) and hard axis ($H \perp c$ axis) and the results are shown in Fig.2 (a) and (b), respectively. In this content, total magnetization is dominated by that of Gd and both figures are scaled by magnetic moment of a Gd atom.

![Figure 2](image2.png)

**Figure 2.** $M$-$H$ curves for Gd=0.8 compound at some temperatures under the magnetic field up to 70,000 Oe. (a) in the easy axis ($H \parallel c$ axis) and (b) in the hard axis ($H \perp c$ axis).
From Fig.2 (a) in easy direction \((H \parallel c\) axis), the Gd magnetic moment, that is, the total magnetization, of this sample increases not linearly but 2-steps likely between 2 K and 15 K, which will be more clearly seen later in section 3.3 at \(x=1\) as spin-flip behaviors. Over temperature of \(T=30\) K, it can be found that the 2-steps like behaviors cannot be observed and disappear. It is remarkable that the Gd magnetic moment saturates well at 2 K and 6 K over the magnetic field of 30,000 Oe and from the value of saturation, the magnetic moment of other element can couple parallel with that of Gd under the conventional assumption that the Gd retains \(7 \mu_B\) since Gd magnetic moment is found to retain over \(7 \mu_B\) from the figure. The candidate of other magnetic element can be Ce and the Ce magnetic moment can be estimated to be \(1.1 \mu_B\) and the magnetic structure in this compound is found to be ferromagnetically coupled Gd ↑ Ce ↑. Taking into account that it is not popular that the Ce magnetic moment saturates completely in Ce122 systems, this estimated value of Ce magnetic moment is considerably valuable.

Fig.2 (b) shows the \(M-H\) curves in hard axis \((H \perp c\) axis) at three temperatures. Since Haen \textit{et al.} have pointed out the unexpected smallness of Ce magnetic moment in hard axis \((H \perp c\) axis), this behavior seems to have attracted little attentions and there have been little reports piled up. From this figure, 2-steps like behavior in the \(M-H\) curves tend to smear out a little by comparing with those in easy axis shown in (a) and the Gd magnetic moment can be clearly observed to saturate over 40,000 Oe at 2 K. Like in easy axis, the Ce is also found to magnetic and retain magnetic moment in hard axis and the magnetic moment of Ce can be estimated to be about \(1.2 \mu_B\) in the same way that in easy axis. In this figure, the total magnetization saturates well over \(H=40,000\) Oe and the value derived out can be reliable. It follows that the Ce magnetic moment in hard axis is nearly identical to that in easy axis and this means that the unexpected smallness of Ce magnetic moment in hard axis does disappear perfectly in Gd content of 0.8. This finding can be the first time to confirm experimentally and this is expected to give an important clue to understand the electronic states of Ce in CeRu2Si2 and Ce122 systems.

3.3. In case of \(x=1\) (i.e. GdRu1.2Si2)

The \(M-H\) curves in Gd content \(x=1\) at some temperatures up to the magnetic field of 90,000 Oe were measured in both directions of easy axis \((H \parallel c\) axis) and hard axis \((H \perp c\) axis) and the results are shown in Fig.3 (a) and (b), respectively.

From Fig.3 (a) in easy axis, 2-steps like spin-flip behaviors can be observed clearly between 2 K and 15 K and roughly 1-step like spin-flip behaviors can be confirmed between 20 K and 40 K. Over 50 K, spin-flip like behavior is found to disappear. The 2-steps like spin-flip behavior have been observed in Nd3Co [15]. Roughly speaking, Nd can be anisotropic since Nd atom retains a component of large angular momentum \(L=6\) and Nd-TM (3d transition metal) including Dy-TM and Ho-TM compounds and alloys are known to be anisotropic and retain generally large coercive force \(H_c\) [16].

![Figure 3](image-url)  
**Figure 3.** \(M-H\) curves for Gd=1 compound at some temperatures under magnetic field up to 90,000 Oe. (a) in the easy axis \((H// c\) axis) and (b) in the hard axis \((H \perp c\) axis).
On the contrary, Gd atom retains no angular momentum (ideally, \(L=0\)) and Gd-TM compounds and alloys retain little and negligible anisotropy and coercive force. That is, considering the above-mentioned, it is not self-evident that the GdRu\(_2\)Si\(_2\) seems to be anisotropic and shows 2-steps spin-flip behaviors like Nd\(_3\)Co. It may be that Gd can retain angular momentum \(L\) effectively in this compound and crystal structure [17]. After all, it is noteworthy that the GdRu\(_2\)Si\(_2\) shows not a simple ferromagnetic behavior but a linear increase in \(M-H\) curve at first and then 2-steps spin-flip behaviors in this compound. This crystal structure can tend to cause the spin-flip behavior in RE elements.

From Fig. 3 (b) in hard axis, roughly speaking, the behaviors in \(M-H\) curves in hard axis resemble to those in easy axis while the spin-flip behaviors in hard axis tend to smear out a little and not sharp in comparison with those in easy axis. Over \(T=50\) K, the behaviors in \(M-H\) curves in both easy and hard axis are considered to be nearly identical.

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References
[1] Steglich F, Aarta J, Bredle C D, Lieke W, Meschede D, Frants W and Schaefer H, 1979 Phys. Rev. Lett. 43 1892
[2] Besnus M J, Kapler P, Lehmann P, Essaihi A, Hamdaoui N, Fischer G, Kappler J P, Meyer A, Pierre J, Haen P and Lejay P 1991 Physica B 171 350
[3] Besnus M J, Kapler P, Lehmann P and Meyer A 1985 Solid State Commun. 55 779
[4] Haen P, Bioud H and Fukuhara T 1999 Physica B 259-261 85
[5] Sekine C, Sakakibara T, Amitsuka H, Miyako Y and Goto T 1992 J. Phys. Soc. Jpn. 61 4536
[6] Murayama S, Sekine C, Yokoyanagi A, Hoshi K and Onuki Y 1997 Phys. Rev. B 56 11092
[7] Flouquet J, Haen P, Raymond S, Aoki D and Knebel G 2002 Physica B 319 251
[8] Lacerda A, Visser de, Haen P, Lejay P, and Flouquet J 1989 Phys. Rev. B 40 8759
[9] Haen P, Flouquet J, Lapierre F, Lejay P and Remenyi G 1987 J Low Temp. Phys. 391
[10] Aoki H, Uji S, Albessard A K and Onuki Y 1993 Phys.Rev.Lett. 71 2110
[11] Okane T, Takeda Y, Yamagami H, Fujimori A, Matsumoto Y, Kimura N, Komatsubara T and Aoki H 2012 Phys.Rev.B 86 125138-1
[12] Sakakibara T, Tayama T, Matsuhira K, Mitani H and Amitsuka H 1995 Phys.Rev.B 51 12030
[13] Mizumaki M, Yano K, Umehara I, Ishikawa F, Sato K, Koizumi A, Sakai N and Muro T 2003 Phys. Rev. B 67 132404-1
[14] Yano K, Ohta T and Sato K 2017 J Phys. Chem. of Solids 104 13
[15] Umehara I, Lu Q, Endo M, Adachi Y and K Sato 1998 J. Phys. Soc. Jpn. 67 4213
[16] Rhyne J J 1979 Amorphous magnetic rare earth alloys (ALLOYS AND INTERMETALLICS vol 2) ed Gschneidner and Eyring (Amsterdam: North-Holland) chapter 16 pp 259-294
[17] Yano K, Okane T, Takeda Y, Yamagami H, Fujimori A, Nishimura K and Sato K 2017 Physica B 515 118