X-ray magnetic circular dichroism study of metamagnetic behaviour in the heavy-fermion system $CeRu_2Si_2$

2003 J. Phys.: Condens. Matter 15 S2171

(http://iopscience.iop.org/0953-8984/15/28/345)

INSTITUTE OF PHYSICS PUBLISHING JOURNAL OF PHYSICS: CONDENSED MATTER

J. Phys.: Condens. Matter **15** (2003) S2171–S2174 PII: S0953-8984(03)62848-1

X-ray magnetic circular dichroism study of metamagnetic behaviour in the heavy-fermion system $CeRu_2Si_2$

T Honma[1], N Kawamura[1], M Suzuki[1], M Mizumaki[1], H Maruyama[2], N Ishimatsu[2], Y Inada[3] and Y Ōnuki[3]

[1] Japan Synchrotron Radiation Research Institute, Mikazuki, Hyogo 679-5198, Japan
[2] Graduate School of Science, Hiroshima University, Higashi-Hiroshima 739-8526, Japan
[3] Graduate School of Science, Osaka University, Toyonaka, Osaka 560-0043, Japan

E-mail: honma@spring8.or.jp

Received 12 November 2002
Published 4 July 2003
Online at stacks.iop.org/JPhysCM/15/S2171

Abstract

Ce unoccupied 5d electronic states of the nonmagnetic heavy-fermion compound $CeRu_2Si_2$ have been investigated by means of x-ray magnetic circular dichroism (XMCD) experiments. We measure the magnetic field dependences of the Ce $L_{2,3}$-edge XMCD of a powder sample with a transmission mode at about 2 K. The magnetic field dependence of the L_2-edge x-ray absorption spectrum shows a slight enhancement of the white line with metamagnetic behaviour, indicating reduction of the c–f hybridization. The L_2 XMCD spectrum shows a single positive lobe, while the L_3 XMCD spectrum consists of a main negative peak and two small negative satellite peaks. The main structure of $CeRu_2Si_2$ is similar to that of $CeRu_2Ge_2$.

1. Introduction

Metamagnetic behaviour of heavy-fermion compounds has been one of the important issues concerning strongly correlated electron systems such as $CeRu_2Si_2$ ($H_c \sim 77$ kOe) [1, 2], URu_2Si_2 ($H_c \sim 400$ kOe) [3] and UPd_2Al_3 ($H_c \sim 180$ kOe) [4]. $CeRu_2Si_2$ is unique among these compounds, in that it is a Pauli paramagnet and has an electronic specific heat coefficient $\gamma \sim 360$ mJ mol^{-1} K^{-2}, while its magnetization exhibits a metamagnetic behaviour with an abrupt nonlinear increase at the field of $H_c \sim 77$ kOe applied along the c-axis below about 10 K.

Over the last decade, a considerable number of studies, such as ones based on the de Haas–van Alphen effect [5], magnetization at very low temperatures [2] and so on, have been made of metamagnetic behaviour. The results of de Haas–van Alphen experiments suggest that the electronic state changes from itinerant to localized with metamagnetic behaviour [5]. But the origin of the metamagnetic behaviour of $CeRu_2Si_2$ is not yet clear.

So far there has been only a small amount of study of the changes of Ce electronic states with metamagnetic behaviour. The valence of Ce in solids has been obtained from Ce L_3-edge x-ray absorption (XA) spectroscopy [6]. On the other hand, x-ray magnetic circular dichroism (XMCD) experiments provide a unique way to obtain information about magnetic polarization in the ferromagnetic state. XMCD is one of the effects in magneto-optics and it is defined as the difference between the amounts of absorption of left and right circularly polarized x-rays. XMCD is a powerful tool for use in investigating electronic states of magnetic atoms because of its element selectivity, electron shell specificity and angular momentum sensitivity.

It is theoretically considered that the ($2p \rightarrow 5d$) $L_{2,3}$-edge XMCD of rare-earth compounds originates in the magnetic moment due to 4f electrons, which induces spin and orbital polarization of 5d electrons through 4f–5d intra-atomic exchange interactions [7]. Therefore, $L_{2,3}$ XMCD is expected to be reflected by the magnetic properties of 4f electrons. Studies of Ce $L_{2,3}$-edge XMCD have been performed experimentally and theoretically for the typical trivalent intermetallic $CeRu_2Ge_2$ compound [8, 9].

Ce $L_{2,3}$-edge XMCD sensitively reflects the response of the unoccupied 4f and 5d electronic states to external fields. Therefore, we have carried out XMCD experiments involving Ce $L_{2,3}$-edge XA spectroscopy at high fields using a superconducting magnet (100 kOe maximum) to clarify the change in the electronic states caused by the metamagnetic behaviour in $CeRu_2Si_2$.

2. Experimental procedure

Ce $L_{2,3}$-edge XA and XMCD spectra of $CeRu_2Si_2$ powder were measured in transmission mode on BL39XU at SPring-8. The left and right circularly polarized x-rays (+ and − helicity) were made by the diamond x-ray phase retarder. We measured the absorption for + helicity and − helicity. In the case where the magnetic field was applied in directions antiparallel to the incident x-ray wavevector, the XA spectroscopy parameter (μt) and XMCD ($\Delta\mu t$) are defined as $\mu t = (\mu_+ t + \mu_- t)/2$ and $\Delta\mu t = \mu_+ t - \mu_- t$, where $\mu_+ t$ and $\mu_- t$ are the absorptions for + and − helicity, respectively. The absorption-edge energy E_0 was determined as the first inflection point of the XA spectrum. The magnetic field dependences of the XA and XMCD spectra were measured up to 100 kOe at about 2 K using a superconducting magnet.

3. Experimental results and discussion

Figure 1(a) shows the Ce L_2-edge XA spectra of $CeRu_2Si_2$, $CeRu_2Ge_2$ and CeO_2. Ce L_2-edge XA spectra of nearly trivalent $CeRu_2Ge_2$ show the strong peak A (white line) and the weak shoulder B. On the other hand, tetravalent CeO_2 shows a double peak A′ and B′ structure. The shift of the absorption-edge energy E_0 from $CeRu_2Ge_2$ to CeO_2 originates in the difference in valence of Ce. The peaks A and A′ are interpreted on the basis of the $2p \rightarrow 5d$ electric dipole (E1) transition with a $4f^1$ configuration, while the weak shoulder B and the peak B′ relate to that with a $4f^0$ configuration [10, 11]. The XA spectrum of $CeRu_2Si_2$ is similar to that of $CeRu_2Ge_2$; therefore $CeRu_2Si_2$ is also close to trivalent. The c–f hybridization of $CeRu_2Si_2$ is slightly stronger than that of $CeRu_2Ge_2$, which is suggested by the only slight enhancement of peak A and reduction of shoulder B for $CeRu_2Si_2$ in comparison with $CeRu_2Ge_2$ [11].

Figure 1(b) shows the difference ΔXAS between the Ce L_2-edge XA spectra (XAS) of $CeRu_2Si_2$ in fields of 60 and 100 kOe and the spectrum in zero field. The change of the XA spectrum is remarkable around the white line and ΔXAS around the white line is

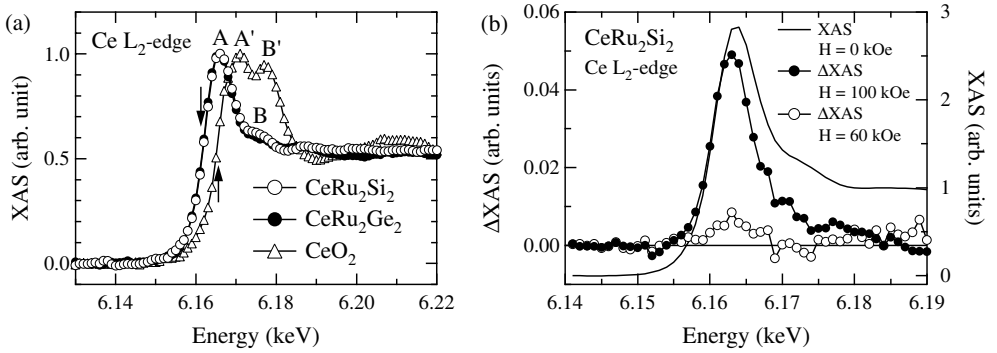

Figure 1. (a) Ce L_2-edge XA spectra of $CeRu_2Si_2$, trivalent $CeRu_2Ge_2$ and tetravalent CeO_2. (b) The difference between the Ce L_2-edge XA spectra in fields of 60 and 100 kOe and the spectrum in zero field for $CeRu_2Si_2$.

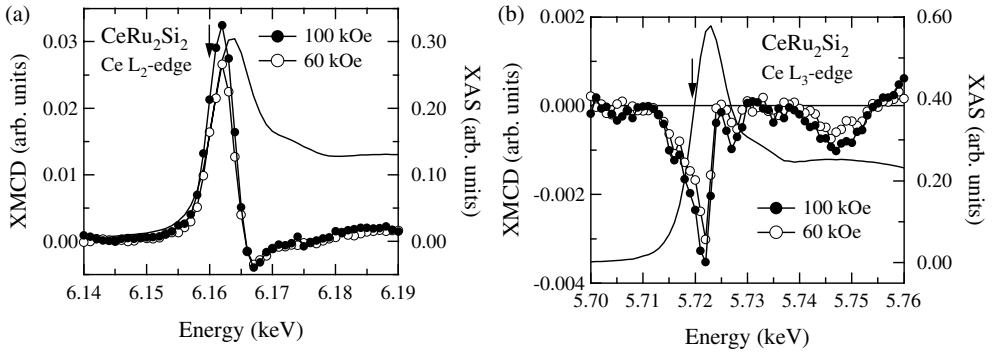

Figure 2. Magnetic field dependences of Ce (a) L_2-edge and (b) L_3-edge XMCD and XA spectra in zero field for $CeRu_2Si_2$.

obviously growing above the crossover field H_c. This result qualitatively suggests that the c–f hybridization in $CeRu_2Si_2$ is decreasing and the 4f state is close to a localized state of $Ce^{3+}(4f^1)$ such as that of $CeRu_2Ge_2$.

The magnetic field dependences of the Ce L_2- and L_3-edge XMCD and XA spectra in zero field for $CeRu_2Si_2$ are shown in figures 2(a) and (b), respectively. The L_2-absorption-edge energy E_0 is 6.160 keV. The L_2 XMCD shows a lobe structure consisting of a main positive peak near the edge and a small negative one at 7 eV above the edge. The L_3-absorption-edge energy E_0 is 5.719 keV. The L_3 XMCD consists of a main negative peak at 3 eV above the edge and small negative peaks at 3 eV below the edge and 8 eV above the edge.

The Ce $L_{2,3}$ XMCD for the trivalent ferromagnetic compound $CeRu_2Ge_2$ was measured by Giorgetti *et al* [8]. The L_2 XMCD spectrum has a single positive lobe at the edge and the L_3 XMCD has a single negative lobe. Although the L_2 XMCD spectrum of $CeRu_2Si_2$ is very similar to that of $CeRu_2Ge_2$, the L_3 XMCD is different from the spectrum of $CeRu_2Ge_2$.

A theoretical study of the Ce $L_{2,3}$ XMCD of $CeRu_2Ge_2$ has been reported by Fukui *et al* [9]. The Ce $L_{2,3}$-edge XMCD of $CeRu_2Ge_2$ is fairly well reproduced by an atomic model, considered as reflecting a spin–orbital interaction and crystal field effects. The XMCD of rare-earth atoms due to the ($2p \rightarrow 4f$) electric quadrupole (E2) transition was also calculated in Fukui's doctoral thesis [12]. The results show a tiny negative lobe below about 5–8 eV

from the peak structure due to the E1 transition in the case of Ce. The contribution of the E2 transition in Ce compounds was ignored in the calculation for $CeRu_2Ge_2$. However, for $CeRu_2Si_2$ a small negative peak is observed at 6 eV below the negative large peak, which most probably originates in the E1 transition. This result suggests that the pre-edge peak is caused by the E2 transition and that the enhancement of the E2 transition in $CeRu_2Si_2$ is a reflection of the large density of states near the Fermi surface in heavy-fermion systems, in spite of the very small E2 transition probability.

4. Summary

Ce L_2- and L_3-edge XA and XMCD spectra of $CeRu_2Si_2$ have been measured with magnetic fields lower and higher than the metamagnetic crossover field H_c. Obvious enhancement of the white line of the Ce L_2-edge XA spectrum due to the metamagnetic behaviour is found, which suggests that the c–f hybridization becomes weak and the 4f state tends to be localized. Although the main structures of the XMCD of $CeRu_2Si_2$ are similar to those for $CeRu_2Ge_2$, a pre-edge peak of the L_3 XMCD for $CeRu_2Si_2$ is observed, which probably originates in the E2 transition. Further theoretical calculations, taking account of the c–f hybridization effect, of the XA and XMCD in strongly correlated electron systems are desired to clarify the origin of the Ce L_3-edge XMCD in $CeRu_2Si_2$.

Acknowledgment

The authors thank K Fukui for discussions.

References

[1] Haen P *et al* 1987 *J. Low Temp. Phys.* **67** 391
[2] Sakakibara T *et al* 1995 *Phys. Rev.* B **51** 12030
[3] Sugiyama K *et al* 1990 *J. Phys. Soc. Japan* **59** 3331
[4] de Visser A *et al* 1992 *Physica* B **179** 81
[5] Aoki H *et al* 1993 *Phys. Rev. Lett.* **71** 2110
[6] Wohlleben D and Röhler J 1984 *J. Appl. Phys.* **55** 1904
[7] Jo T and Imada S 1993 *J. Phys. Soc. Japan* **62** 3721
[8] Giorgetti C *et al* 1993 *Phys. Rev.* B **48** 12732
[9] Fukui K *et al* 1999 *J. Electron Spectrosc. Relat. Phenom.* **104** 67
[10] Soldatov *et al* 1994 *Phys. Rev.* B **50** 5074
[11] Lee C H *et al* 1999 *Phys. Rev.* B **60** 13253
[12] Fukui K 2000 *Doctoral Thesis* Okayama University, Japan