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ARTICLE

Soft x-ray magnetic circular dichroism study on UGe_2

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In order to investigate the electronic and magnetic states of UGe_2 element-specifically, we have performed soft x-ray magnetic circular dichroism experiments at the U $N_{4,5}$ and Ge $L_{2,3}$ absorption edges. We have detected the XMCD signals at both the U and Ge sites and observed clear hysteresis loops in the ferromagnetic (FM) state at T = 5.5 K. From the branching ratio B, it is found that the occupation number of 5f electrons (n^{5f}) in UGe_2 is close to 3. In addition, applying the magneto-optical sum rules analysis to the XMCD spectrum at the U $N_{4,5}$ edges, the ratio of orbital magnetic moment to the spin magnetic moment ($-M_L/M_S$) is estimated to be 2.17. These values of B and $-M_L/M_S$ are comparable compared with the results of other FM superconductors: URhGe and UCoGe.

Keywords: uranium; ferromagnetic superconductors; UGe_2; 5f electron systems; x-ray absorption spectroscopy; magnetic circular dichroism

1. Introduction

In 5f electron system, UGe_2 is the first discovered ferromagnetic (FM) superconductor, which emerges under pressure [1]. UGe_2 crystallizes in the orthorhombic $ZrGa_2$ structure. At ambient pressure, UGe_2 orders ferromagnetically below T_{Curie} = 52 K and the ordered magnetic moment is relative large, M ~ 1.5μ_B along the a-axis. As pressure increases, T_{Curie} decreases. At a higher pressure, eventually the FM state collapses and switches to the paramagnetic state above 1.5 GPa. Around 1.2 GPa, the superconductivity appears below T_{SC} ~0.7 K. Furthermore, there is another characteristic T* ~30 K at ambient pressure as a boundary between FM phase I and FM phase II. As pressure increases, T* connects directly to the superconducting state [2]. In order to investigate the magnetic properties of this compound at the atomic level, we have performed soft x-ray magnetic circular dichroism (XMCD) experiments at the U $N_{4,5}$ and Ge $L_{2,3}$ absorption edges, with changing temperature (T) and magnetic field (H).

2. Experimental

Single crystal samples of UGe_2 were grown by the Czochralski method. The XMCD measurements were carried out at the JAEA contracted beamline BL23SU of SPring-8. A clean surface of the sample was prepared by fracturing *in situ* just before the measurements. T of the sample was controlled between 5.5 K and 90 K using a liquid helium flow cryostat. The external H up to 4 T was applied along the a-axis, the easy axis of magnetization, using a superconducting magnet. The sample was irradiated with the circularly polarized x-ray beam along its magnetization direction (a-axis). X-ray absorption spectra (XAS) were measured by the total electron yield method. The photon helicity can be switched at 1 Hz using combination between a pair of twin-helical undulators of in-vacuum type and five kicker magnets [3]. The XMCD signals were collected by switching the photon helicity at each energy point. The XMCD data displayed in this paper were averaged by reversing H direction in order to correct for experimental artifacts.

3. Results and discussion

Figure 1 (a) shows the XAS spectrum at the U $N_{4,5}$ absorption edges taken at T = 5.5 K and H = 4 T. **Figure 1 (b)** shows the XMCD spectra at several T and H conditions. Here μ^+ (μ^-) refers to the x-ray absorption coefficient for the photon helicity parallel (antiparallel) to the magnetization direction. The XMCD spectrum defined as ($\mu^+ - \mu^-$) is normalized so that the intensity of XAS ($\mu^+ + \mu^-$) from the pre-edge ($h\nu$ = 720 eV) to the peak top at the U N_5 edge ($h\nu$ = 735.8 eV) becomes unity. The shape of the XAS spectra does not depend on T and H (not shown). Although the shape of the XMCD spectra is independent of the experimental conditions, the XMCD intensity increases as T decreases. Compared

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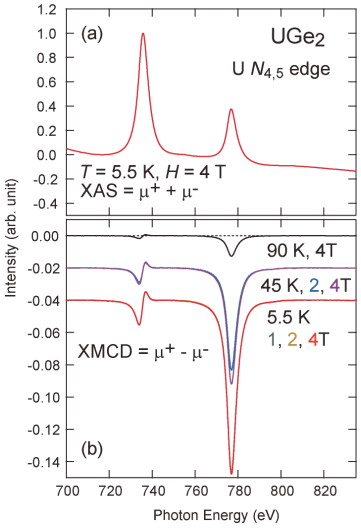

Figure 1. (a) XAS and (b) XMCD spectra of UGe_2 taken at the U $N_{4,5}$ absorption edges. The XMCD spectra were taken at T = 5.5 K, 45 K, and 90 K. At T = 5.5 K, three XMCD spectra measured at H = 1 T, 2 T, and 4 T are completely overlapped.

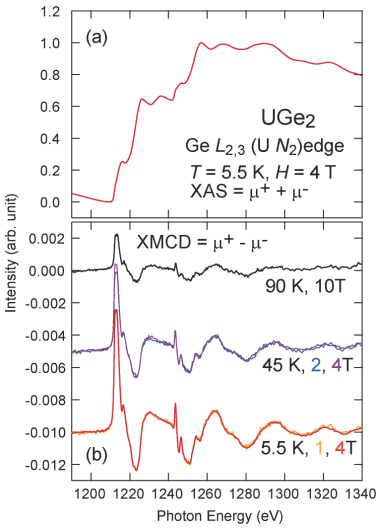

Figure 2. (a) XAS and (b) XMCD spectra of UGe_2 taken at the Ge $L_{2,3}$ absorption edges. The XMCD spectra were taken at T = 5.5 K, 45 K, and 90 K. At T = 5.5 K, two XMCD spectra measured at H = 1 T and 4 T are well overlapped.

with our previous result [4], there is a difference in the spectral shape at the U N_5 edge. As shown in Figure 1 (b), the *s-shaped* structure (a negative peak in lower energy and a positive peak in higher energy), which is commonly seen among uranium compounds, is observed at the U N_5 edge. Before the improvement of our measurement system [3], however, the XMCD spectrum at the U N_5 edge had two negative peaks (main and additional peaks) and no positive peak. In the previous experiment, we have obtained the XMCD signals using μ^+ and μ^- spectra taken by separate energy scans, which could lead to problems of slight energy shift and different back ground between μ^+ and μ^- spectra. The disagreement between the present and the former spectra might be ascribed to such ambiguity in the previous experiment.

Figures 2 (a) and **(b)** show XAS and XMCD spectra at the Ge $L_{2,3}$ absorption edges. The shapes of the XAS and XMCD spectra are not changed with T and H as well. However, the spectral structures are very complicated. In the case of the Ge $L_{2,3}$ edges, the two electron transitions can be considered as $2p \rightarrow 4s$ and $2p \rightarrow 4d$, and there are three different sites of the Ge atoms in the crystalline structure. Moreover, there is the U N_2 absorption edge at hν ~1271 eV. Therefore, it is hard to discuss the electronic states at the Ge sites without theoretical calculation, which is a future plan. Here we would like to emphasize that the XMCD signals are observed clearly at the Ge sites. The existence of magnetic polarization at the Ge sites is consistent with the result observed by the XMCD experiment at the Ge K edge [5].

Figures 3 (a) and **(b)** show the H dependence of the XMCD intensity at the U $N_{4,5}$ (N_5: 734.0 eV, N_4: 776.8 eV) and Ge L_3 (1212.8 eV) edges, namely the element-specific magnetization curves (M-H curves). The hysteresis loop is observed clearly at both the sites. For the U site, the M-H curve at the U N_5 edge shows the same behavior as that at the U N_4 edge. The magnitude of the coercivity is identical between the U and Ge sites. The clear hysteresis is consistent with the result by bulk magnetization measurements.

In order to clarify the electronic states of the U 5f electrons, we have examined a branching ratio B, which corresponds to the relative XAS intensity at the U N_5 edge to that at the U $N_{4,5}$ edges defined as: $B = I_{N5}/(I_{N5} + I_{N4})$. Here, I_{N4} and I_{N5} are the integral values of the XAS spectrum at the U N_4 and N_5 edges, respectively. According to the spin-orbit sum rule based on the relativistic atomic Hartree-Fock calculation in intermediate coupling [6], B can be connected to the occupation number (n^{5f}) of the 5f electrons. The values of B are calculated to be 0.680 and 0.723 for n_{5f} = 2 and 3, respectively. **Figure 4 (a)** shows the integral value (green) of the corrected XAS spectrum (blue) obtained by subtracting a background (a combination of straight lines and arctangent functions). As a result, B is estimated to be 0.71 and is independent of T and H. Here we took the integral values for the I_{N5} and $I_{N5} + I_{N4}$ at hν = 755 eV and 800 eV, respectively. Therefore, n_{5f} in UGe_2 is close to 3. This is consistent with the result by the band-structure calculations within the local density approximation. The calculated n^{5f} by Shick *et al.* has been reported as 2.8 [7]. A recent calculation by Yamagami has given as 2.6 [8]. Compared with other FM superconductors, B is slightly larger in UGe_2 than in URhGe (0.681) [9] and in UCoGe (0.701) [10], which are measured at the U $M_{4,5}$ edges. On the other hand, the early XMCD study for UGe_2 at the U $M_{4,5}$ edges has reported to be 0.66 and suggested that UGe_2 possesses $5f^2$ configuration [11], which is quite different from the results at the U $N_{4,5}$ edges and from the recent results for URhGe and UCoGe. Meanwhile soft x-ray

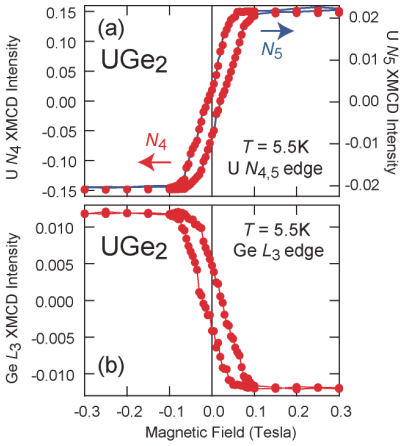

Figure 3. Element-specific magnetization curves (M-H curves) measured at T = 5.5 K. (a) M-H curves taken at the U $N_{4,5}$ edges (N_5: 734.0 eV, N_4: 776.8 eV). The left and right axes are for the N_4 and N_5 edges, respectively. (b) M-H curve taken at the Ge L_3 edge (1212.8 eV).

photoemission experiments have demonstrated that the U 4f core spectra for these FM superconductors show very similar features, suggesting that the electronic states of the U 5f electrons are also close each other [12]. The discrepancy about n^{5f} in UGe_2 between results at the N and M edges is an open question.

Next, we have applied the magneto-optical sum rules to the XMCD spectrum at the U $N_{4,5}$ edges [13, 14]. For N edge absorption, the sum rules are given by:

$$< L_Z >= \frac{14 - n^{5f}}{I_{N5} + I_{N4}} (\Delta I_{N5} + \Delta I_{N4}) \quad (1)$$

$$< S_Z > +3 < T_Z >= \frac{14 - n^{5f}}{2(I_{N5} + I_{N4})} (\Delta I_{N5} - \frac{3}{2} \Delta I_{N4}) \quad (2)$$

Here $<L_Z>$, $<S_Z>$, and $<T_Z>$ are the z-component of the angular momentum, the spin operator, and the intra-atomic magnetic dipole operator, respectively. ΔI_{N5} and ΔI_{N4} correspond to the integral of the dichroic signals at the N_5 and N_4 edges, respectively. Thus the orbital magnetic moment M_L (= $-<L_Z>\mu_B$) and spin magnetic moment M_S (= $-2<S_Z>\mu_B$), and total magnetic moment M_{total} (= M_L + M_S) can be obtained quantitatively if we know the n^{5f} and $<T_Z>$. Although we have not known the n^{5f} and $<T_Z>$ experimentally, the ratios $<T_Z>/<S_Z>$ obtained by the calculation based on the intermediate coupling scheme are 1.16 and 0.62 for the free ions of the $5f^2$ (U^{4+}) and $5f^3$ (U^{3+}) configurations, respectively [15]. Using the ratios and the integral of the XMCD spectrum (taken at T = 5.5 K and H = 4 T) as shown in **Figure 4 (b)**, we have evaluated each value as

Table 1. Analysis using the magneto-optical sum rules [13,14] in the case of U $5f^2$ and $5f^3$ configurations. The ratios $<T_Z>/<S_Z>$ are used the calculated values based on the intermediate coupling scheme [15]. The unit of M_L, M_S, and M_{total} is μ_B/U atom.

n^{5f}	M_L	M_S	M_{total}	$-M_L/M_S$
2	2.03	-0.60	1.43	3.40
3	1.86	-0.86	1.00	2.17

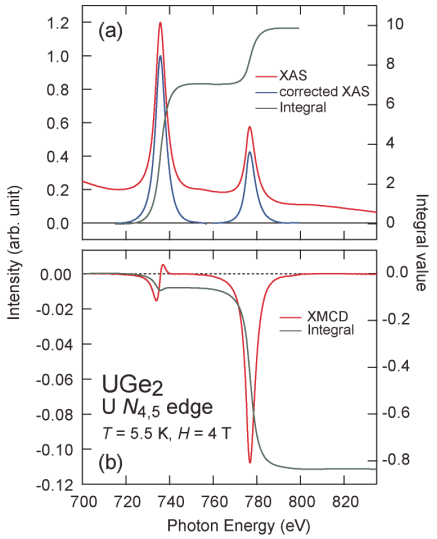

Figure 4. (a) raw XAS spectrum (red) and corrected XAS spectrum (blue) obtained by subtracting a background. The integral of the corrected XAS spectrum (green) is plotted together. (b) XMCD spectrum (red) and its integral (green). The experimental spectra were taken at T = 5.5 K and H = 4 T.

listed in **Table 1** [16]. As seen in the equations (1) and (2), the value of $-M_L/M_S$ is independent of the integral of XAS and of the assumption of n^{5f}. For comparison between the FM superconductors, if the ratio $<T_Z>/<S_Z>$ is kept to be 0.62 (for the case of $5f^3$), the values of $-M_L/M_S$ are 2.17 for UGe_2 (T = 5.5 K and H = 4 T (H //a-axis)), 2.02 for URhGe (T = 2.1 K and H = 2 T (H // c-axis)) [9], and 2.29 for UCoGe (T = 2.1 K and H = 1 T (H // c-axis)) [10]. Here the applied H direction corresponds to each easy axis and the experiments were done in the FM state. These observed values are comparable in spite of the variety of T_C values (UGe_2: T_C = 52 K, URhGe: T_C = 9.5 K, and UCoGe: T_C = 2.7 K), and are less than the value (2.6) for the free U^{3+} ions, implying that the interaction between the U site and other sites is important. As a reference, the value for UCoAl, which is an itinerant metamagnet, is 2.17 (T = 2.1 K and H = 1 T (H // c-axis)) [17] and also is very close. The magnitude of M_{total} (1.0 μ_B/U atom) evaluated by XMCD experiments is less than that obtained by the neutron scattering experiment (1.45 μ_B) [18]. We consider that it is difficult to compare the quantitative magnetic moment because the present analysis is done under simple assumptions. However, It should be noted that the ratio $-M_L/M_S$ for n^{5f} = 3 agrees well between the present XMCD experiment (2.17) and the neutron scattering experiment (2.24).

Finally, the $-M_L/M_S$ of UGe_2 does not change across T^*(~30 K) from T = 5.5 K to 45 K since the spectral shape does not change with the present experimental conditions. In order to investigate the magnetic behavior around T^* in detail, we have planned to promote the systematic T and H dependent XMCD study.

4. Summary

In order to investigate the electronic and magnetic

states of UGe_2, we have performed the soft x-ray magnetic circular dichroism at the U $N_{4,5}$ and Ge $L_{2,3}$ absorption edges. We have detected the clear XMCD signals not only at the U site but also at the non-magnetic Ge sites. Unlike our previous report, the *s-shaped* structure at the U N_5 edge was observed in the present experiment. The value of branching ratio B (0.71) suggests that n^{5f} in UGe_2 is close to 3. In addition, from the analysis using the magneto-optical sum rules under the simple assumption of $5f^3$, the ratio $-M_L/M_S$ is obtained to be 2.17. These values of B and $-M_L/M_S$ of UGe_2 are comparable to those of URhGe and UCoGe.

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[16] The ratio $\langle L_Z\rangle/\langle S_e\rangle$ (where $\langle S_e\rangle=\langle S_Z\rangle+3\langle T_Z\rangle$), which is decided by only the XMCD spectrum and is independent of n^{5f} and $\langle T_Z\rangle$, is -1.52.

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