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Separation of magnetic properties at uranium and cobalt sites in UCoAl using soft x-ray magnetic circular dichroism

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Temperature (T) and magnetic field (H) dependence of the magnetic properties in metamagnetic UCoAl have been investigated using a soft x-ray magnetic circular dichroism (XMCD). In order to extract element-specific magnetic properties at the U and Co sites, the XMCD experiment has been performed at the U 4d-5f (N_{4f}) and Co 2p-3d (L_{2,3}) absorption edges, respectively. Directions of magnetic moments at the U and Co sites have been determined from shapes of the XMCD spectra. The directions of the total magnetic moments at the U and Co sites are parallel to the H direction (c axis), but the direction of the spin magnetic moment at the U site is opposite to that at the Co site. The XMCD intensities at both the U and Co sites at T = 5.5 K increase steeply at H = 0.77 T (H_m), corresponding to the metamagnetic transition. The XMCD intensities do not saturate, even in the field-induced ferromagnetic state above H_m. In addition, the ratio of the increase of the XMCD intensity at the Co site is smaller than that at the U site. From comparison of the H dependence of the XMCD intensities at T = 25 and 5.5 K, we found that the magnetic behavior of the Co atom has a stronger T dependence than that of the U atom.

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I. INTRODUCTION

Uranium compounds display unique and interesting properties, for example, showing a coexistence of superconductivity and magnetism derived from the interaction between the U 5f and other ligand electrons.1 Among the uranium compounds, ternary compounds UTAI (T = Co, Rh and Pt) with hexagonal ZrNiAl-type structure exhibit ferromagnetic ordering. UPtAl and URhAl are ferromagnets with Curie temperature T_C = 43 K (Ref. 2) and 27 K,3 respectively. On the other hand, UCoAl shows a metamagnetic transition (MT) from a paramagnetic ground state to a field-induced ferromagnetic state at low temperature (T).4–7 UCoAl has recently been investigated by resistivity, magnetostriction, and nuclear magnetic resonance experiments from the viewpoint of the quantum critical endpoint.8–10 The MT is strongly anisotropic due to a layered structure with U-Co and Al-Co plane layers stacked consecutively along the c axis. With magnetic field (H) applied along the c axis, the MT occurs at H_m ~ 1 T and T < 15 K with an induced magnetic moment ~0.4 \( \mu_B \), while conventional weak paramagnetic behavior is observed in the perpendicular plane.5–7 The magnetization increases as H/|c|-axis increases up to H = 39 T and does not saturate even above H_m.11 The magnetic behavior of UCoAl, which shows the field-induced ferromagnetic state from the paramagnetic state and unsaturated magnetic moment even at high magnetic fields, is similar to that seen in the itinerant 3d-electron systems YCo_2 and LuCo_2.12,13 Thus, it is important to determine the contribution of the Co atom to the metamagnetism given the Co atom common in these materials. Unlike the high H_m in the 3d-electron systems, for example H_m ~ 70 T for YCo_2, H_m of UCoAl is considerably lower. Furthermore, the MT of UCoAl is observed at ambient pressure and relatively high temperatures. Therefore, UCoAl is a good target for investigating the metamagnetism since the necessary experimental conditions are easily achieved.

Regarding the H dependence of the magnetic moment of UCoAl, there is an interesting result seen in the magnetic Compton scattering (MCS) experiment, which can extract only the component of the spin magnetic moment (M_S).14,15 According to the analysis of the H dependence of the Compton profiles at T = 5 K, the M_S with ~0.1 \( \mu_B \) is observed at H = 1 T (\( \sim H_m \)), but the M_S disappears by H = 3 T.15 Polarized neutron-diffraction (PND) experiments for UCoAl have revealed the H dependence of the magnetic moments at each site element.16,17 However, there is a discrepancy regarding the H dependence of the magnetic moments at the Co site between these PND experiments. The magnetic behavior at each site has not been settled yet. Also there is not enough experimental evidence to understand the disappearance of the M_S observed in the MCS experiment. Therefore, it is necessary to perform a detailed element-specific magnetization curve (M-H curve) measurement.

X-ray magnetic circular dichroism (XMCD) is a powerful technique as an element-specific and electronic orbital selective magnetic probe. In the soft x-ray region, there exist both the Co 2p-3d (L_{2,3}) and the U 4d-5f (N_{4f}) absorption edges. Using soft x-rays, therefore, the magnetic properties at both the U and Co sites can be simultaneously investigated in the same experiment only by tuning the photon energy to each absorption edge. In addition, since the XMCD intensity is proportional to the magnitude of the magnetic moments, the element-specific M-H curve measurement can be done by investigating the H dependence of the XMCD intensity.18,19

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As for the magnetic properties of the U 5f electrons of UCoAl, the H-dependent XMCD experiments at the U 3d-5f (M4,5) absorption edge have already been carried out in the hard x-ray region, but the magnetic properties of the Co 3d electrons can never be investigated directly in the hard x-ray region.

In this paper, we have carried out the H- and T-dependent XMCD experiments of UCoAl at the U N4,5 and the Co L2,3 edges in order to investigate the magnetic properties at both the U and Co sites. We decide the directions of the magnetic moments (total magnetic moment: M\text{total}, M\text{S}, and M\text{L}) of the U 5f and Co 3d electrons from the shape of the XMCD spectrum. We discuss the H and T dependence of the magnetic moments of the U 5f and Co 3d electrons using the element-specific M-H curve measurement.

**II. EXPERIMENT**

We used single crystals grown using the Czochralski method in a tetra-arc furnace. The XMCD experiments at the U N4,5 and the Co L2,3 edges were carried out at the beam line BL23SU of SPring-8. The external H up to 7 T was applied to the sample along the c axis, the easy axis of magnetization, using a superconducting magnet. The incident circularly polarized x-ray irradiated the sample along the c axis. X-ray absorption spectroscopy (XAS) spectra were obtained by the total electron yield method. In the XMCD end station, the helicity switching of circular polarization is done at 1 Hz using a twin-helical undulator of the in-vacuum type. XMCD signals were obtained by the helicity switching at each energy. In the element-specific M-H curve measurements, the U N5 and Co L2 edges were chosen due to an overlap of the spectra at the U N5 and Co L3 edges. A clean surface of the sample was obtained by fracturing in ultrahigh vacuum. The sample was cooled to T = 5.5 and 25 K using a liquid-helium cryostat.

**III. RESULTS AND DISCUSSION**

Figures 1(a) and 1(b) show the XAS and XMCD spectra of UCoAl at the U N4,5 and Co L2,3 edges. The spectra were obtained in the field-induced ferromagnetic state at T = 5.5 K and H = 7 T. Here, \(\mu^+ (\mu^-)\) refers to the x-ray absorption coefficient for the photon helicity parallel (antiparallel) to the magnetization direction. The inset of Fig. 1(a) shows the expanded plots of the \(\mu^+\) and \(\mu^-\) absorption spectra around \(\hbar \nu = 778\) eV. The intensity of the XMCD spectrum defined as \((\mu^+ - \mu^-)\) is normalized so that the peak intensity of XAS (\(\mu^+ + \mu^-\)) at the U N5 edge (\(\hbar \nu = 736.6\) eV) becomes 1 [Fig. 1(a)]. Although the two pairs of the spin-orbit splitting exist in the photon energy region, namely, the U N4,5 and Co L2,3 edges, only three peaks are observed in the XMCD spectrum due to the spectra at the Co L3 and U N4 edges overlapping around \(\hbar \nu = 778\) eV. The XMCD signals of the two nonequivalent Co sites cannot be distinguished in the present XMCD experiment.

Figures 2(a) and 2(b) show the H dependence of the XMCD spectra at T = 5.5 and 25 K, respectively. The normalization is done in the same way as that in Fig. 1. At T = 5.5 K, a drastic growth of XMCD intensity from \(H = 0.5\) to 1 T is observed in all of the absorption edges, corresponding to the MT from the paramagnetic state to the field-induced ferromagnetic state.

A monotonous increase of the XMCD intensity from \(H = 1\) to 7 T is consistent with the previous result from conventional magnetization measurement. The inset of Fig. 2(a) displays the enlarged spectra at the U N5 edge. The intensities are normalized again so that the negative peak of the XMCD spectra at \(h\nu = 735.0\) eV becomes \(-1\) in order to facilitate a comparison of the spectral shape. The XMCD spectrum at the U N5 edge has the asymmetric “s” shape of a two-peak structure with a negative and positive sign. Theory predicts that the shape of the XMCD spectra at the U N5 and M5 edges is sensitive to the valence of the U atom. In the previous XMCD study using hard x-rays, the spectral shape at the U M5 edge changed with \(H\). The XMCD spectra at the U M5 edge have the “s” shape in magnetic fields above \(H = 1\) T, but the positive peak disappears and becomes a single negative peak at \(H = 0.7\) T. However, the authors mentioned that the change of the spectral shape might be ascribed to the small signal-to-noise ratio of the measurement at the U M5 edge. In the present measurements, the “s” shape at the U N5 edge survives definitely even at \(H = 0.5\) T and there is no change of the shape at \(H = 0.5, 1, 3, 5,\) and 7 T, as shown in the inset of Fig. 2(a). Also, we have confirmed that the XMCD spectra at \(T = 25\) K and \(H = 2\) and 7 T have the “s” shape [Fig. 2(b)].

In the present XMCD study, it is difficult to estimate a quantitative value of the magnetic moments by applying the sum rules due to the overlap of the spectra at the U N4 and the Co L3 edges. However, the directions of the M5 and M3 can be deduced only from the shape of the XMCD spectrum.
According to the previous XMCD studies on various uranium compounds, it is well known that the XMCD spectrum at the U $N_4$ edge has a symmetric line shape with a negative sign.\textsuperscript{22,26,27} In fact, the symmetric line shape is observed in the XMCD spectrum of UCoAl at the U $N_4$ edge.\textsuperscript{20} Therefore, we can deduce that the XMCD spectrum at the U $N_4$ edge of UCoAl has a symmetric structure with a negative sign. In Fig. 3(a), the red dashed line shows the deduced XMCD spectrum of UCoAl at the U $N_4$ edge.\textsuperscript{28} Here the peak intensity ratio of $N_4/N_5$ of the XMCD spectrum is consistent with the ratio of $M_4/M_5$ at the U $M_{4,5}$ edge within an error of 10%.\textsuperscript{20} We find that the XMCD intensity around $h\nu \sim 770$–$776$ eV consists of the intensity at the U $N_4$ edge mainly. Thus the integral of the XMCD intensity at the Co $L_3$ edge is a negative value because the XMCD spectrum around $h\nu = 778$ eV has a large residual intensity. The integral of the XMCD intensity at the Co $L_2$ edge is indicated to be a positive value, as shown in Fig. 3(a). The absolute value of the integral at the Co $L_3$ edge is larger than that at the Co $L_2$ edge, suggesting that a finite $M_L$ at the Co site exists.\textsuperscript{29} On the other hand, the integrals of the XMCD intensity at both the U $N_4$ and $N_5$ edges denote negative values. As compared with the XMCD spectrum of UF$_2$ at the U $N_{4,5}$ and Fe $L_{2,3}$ edges, the spectral configuration between the U and Co sites in UCoAl is the same as that between the U and Fe sites in UF$_2$.\textsuperscript{27} As a result, we conclude that the directions of the magnetic moments of the U 5$f$ and the Co 3$d$ electrons are shown in Fig. 3(b). The $M_{\text{total}}$ directions at both the U and Co sites turn parallel to the direction of $H$ (c axis). At the U site, the $M_L$ is parallel to the $M_{\text{total}}$ direction and the $M_S$ is antiparallel to $M_{\text{total}}$. On the other hand, both $M_S$ and $M_d$ directions are parallel to the $M_{\text{total}}$ direction at the Co site. Consequently, the $M_S$ at the U site is antiparallel to the $M_S$ at the Co site. The conclusion is consistent with the results obtained from the PND experiments.\textsuperscript{16,17} Since there is no change in the spectral configuration of the XMCD spectra with $H$ and $T$, the relation of the magnetic moments is conserved even in the
paramagnetic state at \( H = 2, 7 \) T and \( T = 25 \) K [Fig. 2(b)], as well as at \( H = 0.5, 1, 3, 5, 7 \) T and \( T = 5.5 \) K [Fig. 2(a)].

In order to investigate the detailed \( H \) dependence of the XMCD intensity at the U and Co sites, the M-H curve measurements were performed by changing \( H \) and \( T \) and by tuning the photon energies to the U \( N_5 \) and Co \( L_2 \) edges. Figure 4 shows the \( T \) dependence of the M-H curves at the U \( N_5 \) (\( hν = 735.0 \) eV) and Co \( L_2 \) (\( hν = 794.5 \) eV) edges. The M-H curves were measured along a loop pathway (\( H = 7 \) T \( \rightarrow \) -7 T \( \rightarrow \) 7 T). The XMCD intensities plotted at the magnetic fields in Fig. 4 are obtained by averaging the XMCD signals at the corresponding \( \pm H \). The M-H curves at the U \( N_5 \) and Co \( L_2 \) edges are plotted on the left and right vertical axes, respectively. The inset of Fig. 4 shows the M-H curves taken at \( hν = 715 \) eV. Since there are no absorption edges at \( hν = 715 \) eV, the M-H curves are flat and independent of \( H \) and \( T \). These M-H curves at \( hν = 715 \) eV can be understood as the present precision of the M-H curve measurements. At \( T = 25 \) K, the XMCD intensities at the U and Co sites increase gently as \( H \) increases (Fig. 4). When the intensities of the M-H curve at \( T = 25 \) K are normalized, it can be found that the curvature of the M-H curve at the U site is approximately the same as that at the Co site. This indicates that the magnetic moments at both sites have the same \( H \) dependence in the paramagnetic state.

As \( T \) goes down to \( T = 5.5 \) K, the M-H curves show steep jumps at both the U and Co sites at \( H = 0.77 \) T (\( H_m \)), where the MT occurs. Here the value of the \( H_m \) is estimated by the second derivative of the M-H curve. According to the bulk magnetization measurement, the hysteresis at \( T = 5.5 \) K is expected to be \( \sim 0.02 \) T (Ref. 8); however, it was not observed clearly within the precision. The XMCD intensity at \( T = 5.5 \) K is plotted on the common scale at \( T = 25 \) K. Above \( H_m \), the XMCD intensities at both the U and Co sites increase monotonically as \( H \) increases. Unlike the M-H curves at \( T = 25 \) K, the ratio of the increase of the XMCD intensity at the U site is larger than that at the Co site in the field-induced ferromagnetic state. In order to see the difference in the slope easily, we show the dashed straight lines obtained by fitting the M-H curves between \( H = 1 \) and 7 T. As \( H \) increases from \( H = 1 \) to 7 T, the XMCD intensities at the U and Co sites increase by the factors of \( \sim 1.9 \) and \( \sim 1.3 \), respectively. Therefore, we propose that the opposite directions of \( M_S \) [Fig. 3(b)] and the different magnetic response at each site [Fig. 4] give plausible reason for the cancellation of the \( M_S \) observed in the MCS experiment.14

From the bulk magnetization measurement,3 however, the bulk magnetic moment (\( M_{\text{bulk}} \)) at \( T = 5 \) K increases by a factor of \( \sim 1.3 \) from \( H = 1 \) to 7 T (from 0.37 to 0.48 \( \mu_B \), respectively), which is smaller than the ratio of the increase of the XMCD intensity at the U site. Since the directions of the \( M_{\text{total}} \) at the U and Co sites are the same [Fig. 3(b)], the ratio of the increase of the \( M_{\text{bulk}} \) cannot be explained only by the \( M_{\text{total}} \) of the U 5f and Co 3d electrons, implying that other magnetic contributions to the \( M_{\text{bulk}} \) exist in UCoAl. Indeed, the PND experiment in Ref. 17 has suggested the existence of magnetic moments at the Al site and/or of the other conduction electrons. In order to compare the \( H \) dependence of the magnetic moments at the U and Co sites in the field-induced ferromagnetic state, the ratio of the increase of magnetic moments from \( H = 1 \) to 7 T (\( R_{\text{H–T}} \)) observed in the XMCD20 and PND experiments16,17 is summarized in Table I. According to the XMCD experiments using the hard x-ray, the \( R_{\text{H–T}} \) at the U site is estimated to be \( \sim 2.2 \) at \( T = 10 \) K from the \( H \) dependence of the XMCD intensity,20 which is close to that observed in the present XMCD experiment. There are two reports about the \( H \) dependence of the magnetic moments by the PND experiments.16,17 In the case of the PND experiments, the magnetic moment at the Co site is regarded as the sum of the magnetic moments of the two nonequivalent Co sites. One has been performed at \( H = 1.7 \) and 5 T and at \( T = 5 \) K in Ref. 16 and the \( R_{\text{H–T}} \) is deduced from linear extrapolation.

### Table I. \( R_{\text{H–T}} \) at the U and Co site obtained from the XMCD and PND experiments.

| Experiment       | U site | Co site |
|------------------|--------|---------|
| Present XMCD exp. | 1.9 ± 0.1 | 1.3 ± 0.1 |
| XMCD exp. (U \( M_{5.5} \) edge) | 2.2 ± 0.1 | 1.3 |
| PND exp.a | 1.3 | 1.3 |
| PND exp.b | 1.2–1.3 | 0.9–1.2 |

a In the case of the PND experiments, it is the sum of the two nonequivalent Co sites.
b Ref. 20. At \( T = 10 \) K.

c Ref. 16. At \( T = 5 \) K, \( H = 1.7 \) and 5 T. The \( R_{\text{H–T}} \) is deduced from linear extrapolation.
d Ref. 17. At \( T = 2 \) K, \( H = 1 \) and 8 T. The \( R_{\text{H–T}} \) is deduced from linear interpolation.
The $R_{\text{IT}-7T}$ at the U site is $\sim 1.3$ and is nearly equal to that at the Co site. The other has been done at $H = 1$ and $8 \ T$ at $T = 2 \ K$ in Ref. 17 and the $R_{\text{IT}-7T}$ is deduced from linear interpolation. They have proposed several methods to estimate the magnitude of magnetic moments from the PND data. Thus, the $R_{\text{IT}-7T}$ depends on the analysis methods with a large error. As a result, the $R_{\text{IT}-7T}$ is $\sim 1.2\text{–}1.3$ and $\sim 0\text{(decrease)}\sim 1.2$ at the U and Co sites, respectively. There is a discrepancy about the $H$ dependence of the magnetic moments at the Co site between these PND experiments, although the $R_{\text{IT}-7T}$ at the U site seems to be comparable to that from the bulk magnetization measurement. In Ref. 17, the $R_{\text{IT}-7T}$ at the Co site is small compared with that at the U site, which is different from the result observed in Ref. 16. Therefore, the relation of the magnetic behavior between the U and Co sites ($R_{\text{IT}-7T; U > Co}$), which is observed in the present XMCD experiment, is in qualitative agreement with that reported in Ref. 17.

Finally, it should be noted that the magnetic behavior of the Co atom has a stronger $T$ dependence than that of the U atom. The slope of the M-H curve at each element site can be regarded approximately as the element-specific magnetic susceptibility ($\chi_{\text{site}}^E$). Here we have obtained $\chi_{\text{site}}^E$ by fitting the M-H curve at $T = 25 \ K$ above $H = 3 \ T$, and $\chi_{\text{site}}^E$ corresponds to the slope of the dashed line in Fig. 4. From $T = 25$ to 5.5 $K$, the slope of the M-H curve is decreased at both the U and Co sites, i.e., $\chi_{\text{site}}^{\text{U}}/\chi_{\text{site}}^{\text{Co}} < 1$ and $\chi_{\text{site}}^{\text{Co}}/\chi_{\text{site}}^{\text{U}} < 1$. By comparing the U and Co sites, we have found the relation $\chi_{\text{site}}^{\text{Co}}/\chi_{\text{site}}^{\text{U}} < \chi_{\text{site}}^{\text{U}}/\chi_{\text{site}}^{\text{Co}}$, indicating that the slope of the M-H curve becomes smaller at the Co site than at the U site as $T$ goes down. The result suggests that it is important to clarify the contribution of the Co atom to the MT of UCoAl. A detailed $T$ dependence of the element specific M-H curve will provide crucial information about a mechanism of the MT.

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The XMCD spectra at the U$^{4+}$ edge which have been measured so far are very similar so it is difficult to distinguish. The line shape of the XMCD spectrum at the U$^{4+}$ edge can be reproduced well by the Voigt function. The red dashed line in Fig. 3(a) is based on the Lorentzian function with the peak position 778.3 eV and the full width at half height 5.3 eV.

Assuming that the x-ray absorption occurs through the electric dipole transitions, the $M_L$ is proportional to the integral of the XMCD intensity over the $L_3$ and $L_2$ edges. When the sign of the integral is negative (positive), the $M_L$ turns parallel (antiparallel) to the external magnetic field.