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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 $4d-5f$ ($N_{4,5}$) 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 ($T = \text{Co, Rh and Pt}$) 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 $H_m \sim 1$ T and $T < 15$ K with an induced magnetic moment $\sim 0.4 \mu_B$, 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 YCo₂ and LuCo₂.^{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 $H_m \sim 70$ T for YCo₂, 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 $\sim 0.1 \mu_B$ is observed at $H = 1$ T ($> H_m$), but the M_S disappears by $H = 3$ T.¹⁴ Polarized neutron-diffraction (PND) experiments for UCoAl have revealed the H dependence of the magnetic moments at each element site.^{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 $4d-5f$ ($N_{4,5}$) 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}

As for the magnetic properties of the U $5f$ electrons of UCoAl, the H -dependent XMCD experiments at the U $3d$ - $5f$ ($M_{4,5}$) absorption edge have already been carried out in the hard x-ray region,²⁰ 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 $N_{4,5}$ and the Co $L_{2,3}$ 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_{total} , M_S , and M_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.⁵ The XMCD experiments at the U $N_{4,5}$ and the Co $L_{2,3}$ edges were carried out at the beam line BL23SU of SPring-8.²¹ 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.²² XMCD signals were obtained by the helicity switching at each energy. In the element-specific M-H curve measurements, the U N_5 and Co L_2 edges were chosen due to an overlap of the spectra at the U N_4 and Co L_3 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 $N_{4,5}$ and Co $L_{2,3}$ edges. The spectra were obtained in the field-induced ferromagnetic state at $T = 5.5$ K and $H = 7$ T. Here, μ^+ (μ^-) 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 μ^+ and μ^- absorption spectra around $h\nu = 778$ 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 N_5 edge ($h\nu = 736.6$ eV) becomes 1 [Fig. 1(a)]. Although the two pairs of the spin-orbit splitting exist in the photon energy region, namely, the U $N_{4,5}$ and Co $L_{2,3}$ edges, only three peaks are observed in the XMCD spectrum due to the spectra at the Co L_3 and U N_4 edges overlapping around $h\nu = 778$ 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.

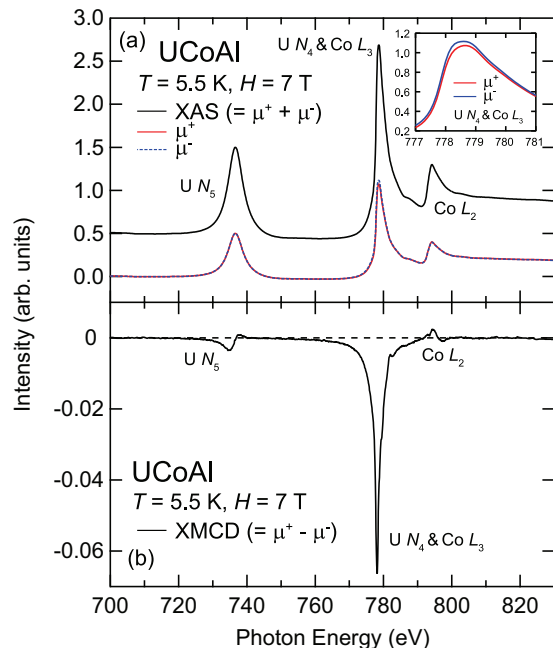


FIG. 1. (Color online) The XAS and XMCD spectra of UCoAl at the U $N_{4,5}$ and the Co $L_{2,3}$ edges at $T = 5.5$ K and $H = 7$ T. The U N_4 edge overlaps with the Co L_3 edge. (a) The μ^+ (μ^-) refers to the absorption spectrum for the photon helicity parallel (antiparallel) to the magnetization direction. The XAS spectrum is defined as $\mu^+ + \mu^-$. Inset: The magnified figure of the μ^+ and μ^- absorption spectra around $h\nu = 778$ eV. (b) The XMCD spectrum defined as $\mu^+ - \mu^-$. These spectra are normalized so that the intensity of the XAS ($\mu^+ + \mu^-$) spectrum at the U N_5 edge ($h\nu = 736.6$ eV) becomes 1.

A monotonous increase of the XMCD intensity from $H = 1$ to 7 T is consistent with the previous result from conventional magnetization measurement.⁵ The inset of Fig. 2(a) displays the enlarged spectra at the U N_5 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 N_5 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 N_5 and M_5 edges is sensitive to the valence of the U atom.²³ In the previous XMCD study using hard x-rays, the spectral shape at the U M_5 edge changed with H .²⁰ The XMCD spectra at the U M_5 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 M_5 edge. In the present measurements, the “s” shape at the U N_5 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^{24,25} due to the overlap of the spectra at the U N_4 and the Co L_3 edges. However, the directions of the M_L and M_S can be deduced only from the shape of the XMCD spectrum.

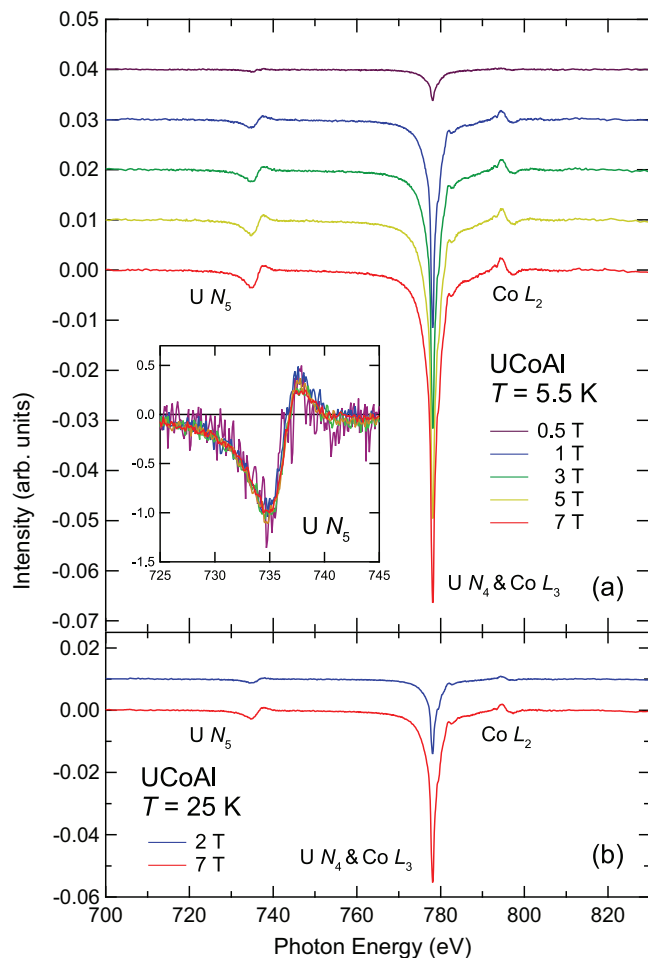


FIG. 2. (Color online) The H dependence of the XMCD spectra of UCoAl at the U $N_{4,5}$ and Co $N_{2,3}$ edges. These spectra are normalized as in Fig. 1. (a) At $T = 5.5$ K. Inset: The magnified figure of the XMCD spectra at the U N_5 edge. These spectra at each magnetic field are normalized so that the negative peak of the XMCD spectra at $h\nu = 735.0$ eV becomes -1 . (b) At $T = 25$ K.

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.^{22,26,27} In fact, the symmetric line shape is observed in the XMCD spectrum of UCoAl at the U M_4 edge.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.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%.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.²⁹ On the other hand,

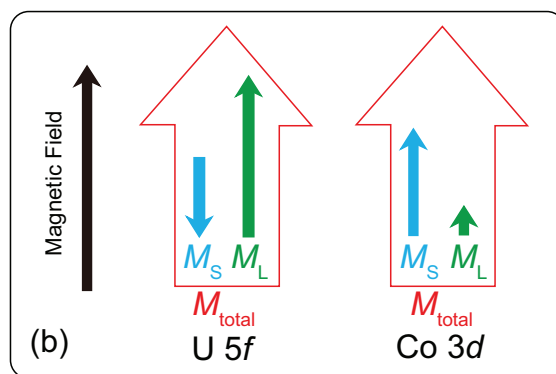
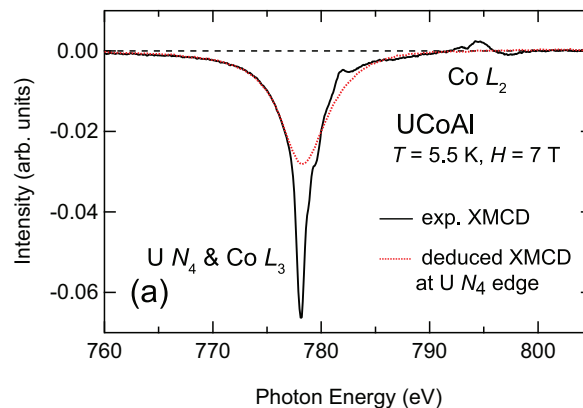


FIG. 3. (Color online) Decision of the direction of the magnetic moments at the U and Co sites. (a) The expanded plot of the XMCD spectrum around the U N_4 and Co L_3 edges. The red dashed line shows the deduced XMCD spectrum at the U N_4 edge. See text for details. (b) A schematic picture of the magnetic moments (total magnetic moment M_{total} , spin magnetic moment M_S , and orbital magnetic moment M_L) deduced from the shape of the XMCD spectrum at the U $N_{4,5}$ and Co $L_{2,3}$ edges. The size of the arrows is drawn schematically because the magnitude of magnetic moments cannot be estimated quantitatively. However, the U $5f$ moments are drawn in accordance with the result from the XMCD experiment at the U $M_{4,5}$ edge²⁰ so that the ratio $-M_L/M_S$ of the U $5f$ electrons at $H = 7$ T and $T = 10$ K is estimated to be 1.95 in the case of the U $5f^3$ configuration.

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 UFe₂ 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 UFe₂.²⁷ As a result, we conclude that the directions of the magnetic moments of the U $5f$ and the Co $3d$ electrons are shown in Fig. 3(b). The M_{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_{total} direction and the M_S is antiparallel to M_{total} . On the other hand, both M_S and M_L directions are parallel to the M_{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.^{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

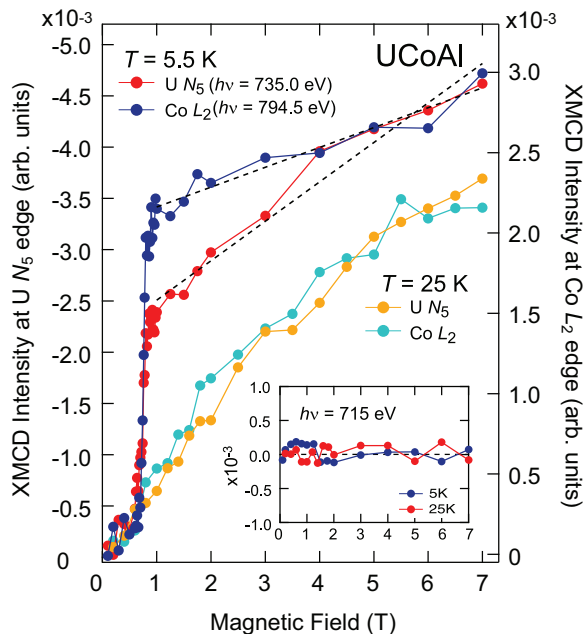


FIG. 4. (Color online) The H dependence of the XMCD intensities (M-H curve) at the U N_5 ($h\nu = 735.0$ eV) and Co L_2 ($h\nu = 794.5$ eV) edges at $T = 5.5$ and 25 K. The dashed straight lines are obtained by fitting the M-H curves between $H = 1$ and 7 T. These straight lines are guides to the eye. Inset: M-H curves taken at $h\nu = 715$ eV, where there is no absorption edge, indicating the precision of the M-H curve measurements.

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 the Co L_2 edges. Figure 4 shows the T dependence of the M-H curves at the U N_5 ($h\nu = 735.0$ eV) and Co L_2 ($h\nu = 794.5$ 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 $h\nu = 715$ eV. Since there are no absorption edges at $h\nu = 715$ eV, the M-H curves are flat and independent of H and T . These M-H curves at $h\nu = 715$ 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

TABLE I. R_{1T-7T} at the U and Co site obtained from the XMCD and PND experiments.

Experiment	U site	Co site ^a
Present XMCD expt.	1.9 ± 0.1	1.3 ± 0.1
XMCD expt. (U $M_{4,5}$ edge) ^b	2.2 ± 0.1	
PND expt. ^c	1.3	1.3
PND expt. ^d	1.2–1.3	0.9–1.2

^aIn the case of the PND experiments, it is the sum of the two nonequivalent Co sites.

^bRef. 20. At $T = 10$ K.

^cRef. 16. At $T = 5$ K, $H = 1.7$ and 5 T. The R_{1T-7T} is deduced from linear extrapolation.

^dRef. 17. At $T = 2$ K, $H = 1$ and 8 T. The R_{1T-7T} is deduced from linear interpolation.

bulk magnetization measurement, the hysteresis at $T = 5.5$ K is expected to be ~ 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 ~ 1.9 and ~ 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,⁵ however, the bulk magnetic moment (M_{bulk}) at $T = 5$ K increases by a factor of ~ 1.3 from $H = 1$ to 7 T (from 0.37 to 0.48 μ_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_{total} at the U and Co sites are the same [Fig. 3(b)], the ratio of the increase of the M_{bulk} cannot be explained only by the M_{total} of the U $5f$ and Co $3d$ electrons, implying that other magnetic contributions to the M_{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_{1T-7T}) observed in the XMCD²⁰ and PND experiments^{16,17} is summarized in Table I. According to the XMCD experiments using the hard x-ray, the R_{1T-7T} at the U site is estimated to be ~ 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_{1T-7T} is deduced from linear extrapolation.

The R_{1T-7T} at the U site is ~ 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_{1T-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_{1T-7T} depends on the analysis methods with a large error. As a result, the R_{1T-7T} is $\sim 1.2-1.3$ and $\sim 0.9(\text{decrease})-1.2$ 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_{1T-7T} at the U site seems to be comparable to that from the bulk magnetization measurement.⁵ In Ref. 17, the R_{1T-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_{1T-7T} ; $U > \text{Co}$), 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 (χ_T^{site}). Here we have obtained $\chi_{25\text{K}}^{\text{site}}$ by fitting the M-H curve at $T = 25$ K above $H = 3$ T, and $\chi_{5.5\text{K}}^{\text{site}}$ 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., $\chi_{5.5\text{K}}^{\text{U}}/\chi_{25\text{K}}^{\text{U}} < 1$ and $\chi_{5.5\text{K}}^{\text{Co}}/\chi_{25\text{K}}^{\text{Co}} < 1$. By comparing the U and Co sites, we have found the relation $\chi_{5.5\text{K}}^{\text{Co}}/\chi_{25\text{K}}^{\text{Co}} < \chi_{5.5\text{K}}^{\text{U}}/\chi_{25\text{K}}^{\text{U}}$, 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.

IV. SUMMARY

We have performed the element-specific investigation of the magnetic properties in metamagnetic UCoAl via the XMCD experiment at the U $N_{4,5}$ and the Co $L_{2,3}$ edges. We have succeeded in independently extracting the magnetic behavior at the U and the Co sites. The directions of the magnetic moments at the U and Co sites are decided from the shape of the XMCD spectrum. The directions of the total magnetic moment M_{total} of both the U $5f$ and Co $3d$ electrons turn parallel to the direction of the external H (c axis). But the spin moment M_S of the U $5f$ electrons turns in the opposite direction of that of the Co $3d$ electrons. From the element-specific M-H curve measurement, the sharp development in the XMCD intensity at both the U and Co sites is clearly observed at $T = 5.5$ K and $H_m = 0.77$ T, corresponding to the MT. The XMCD intensity at both sites does not saturate even above H_m . In the paramagnetic state ($T = 25$ K), the curvature of the M-H curve is approximately the same at both the U and Co sites. However, in the field-induced ferromagnetic state above H_m ($T = 5$ K), the ratio of the increase of magnetic moments at the Co site becomes smaller than that at the U site. This fact indicates that the magnetic behavior of the Co atom has a stronger T dependence than that of the U atom.

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- ²⁸The XMCD spectra at the U N_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 N_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.
- ²⁹Assuming 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.