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Magnetic Compton of USe Single crystal

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The magnetic Compton scattering experiment of USe single crystal was performed . The spin moment μ_s of USe was determined from magnetic Compton profile (MCP) and the orbital moment μ_L was deduced by combining the μ_S with the result of magnetization measurement . Furthermore , the spin moments of USe were decomposed into 5f electron component of uranium and conduction-electron-like component by fitting with calcurated Compton profiles from ralativistic Dirac-Hartree-Fock wave functions . Decomposed components for USe are discussed in terms of Russel-Saunders coupling models .

KEYWORDS: USe , Magnetic Compton profile (MCP) , spin moment μ_s , orbital moment μ_L

1 Introduction

Recently , much attention has been paid to the magnetic and electronic properties of actinide compounds from both theoretical and experimental points of view . In order to understand these properties , it is essential to study the behavior of 5f electrons which may have a wide range of characteristic features from " localized " to " itinerant " in nature . From these points of view , uranium monochalcogenides (US , USe and UTe) with simple NaCl type structure have been extensively studied . These compounds show ferro-magnetic properties with relatively higher transition temperature (T_c=180 , 160 , 104K , respectively) and show strong magnetocrystalline anisotropy along the <111> direction [1]. Furthermore , these compounds show large polar Kerr rotation angle[2], which is due to the orbital polarization [3] . So it is valuable to investigate the behavior of 5f electron in order to explain the origin of these phenomena .

Magnetic Compton scattering experiment reflects only spin moment μ_s [4][5] . So by combining with the magnetization , the orbital moment μ_L can be deduced .

In a previous paper , we reported the magnetic Compton scattering experiment and the separated μ_S and μ_L of UTe . In this paper , we report the magnetic Compton profile of USe and separate the magnetic moment to orbital part and spin part in USe .

2 Experimental

2.1 Magnetic Compton profile

The Compton profile , $J(P_z)$, is the projection of the electron moment density , $n(\vec{P})$, along the scattering vector which is conventionally defined as the z-direction ,

$$J(P_z) = \int\int n(\vec{P}) dP_x dP_y. \tag{1}$$

This expression is for unpolarized photons and assumes the impulse approximation , i.e. , that the energy of recoil electrons is sufficiently high , compared with the binding energy for the final state to be treated as the plane wave . The electron momentum P_z is then given by the following equation

$$\frac{P_z}{mc} = \frac{E_2 - E_1 \pm \left(\frac{E_1 E_2}{mc^2}\right)(1 - \cos\theta)}{\sqrt{E_1^2 + E_2^2 - 2E_1 E_2 \cos\theta}}, \tag{2}$$

where θ is the scattering angle , E_1 and E_2 are the incident and the scattered X-ray energies , respectively , m is the electron mass , and c is the velocity of light . When polarized photons are used , the scattering cross section can be written as

$$\frac{d^2\sigma}{d\Omega dE_2} = r_0^2 \frac{m}{2\hbar K} \left(\frac{E_2}{E_1}\right) \left[f_1 J(P_z) + f_2 \frac{E_1}{mc^2} P_c S(\alpha) J_{mag}(P_z) \right], \tag{3}$$

where

$$f_1 = 1 + \cos^2\theta + \frac{E_1 - E_2}{mc^2}(1-\cos\theta) + P_l \sin^2\theta, \quad (4)$$

$$f_2 = -(1-\cos\theta), \quad (5)$$

and

$$S(\alpha) = \sigma\left(\cos\alpha\cos\theta + \frac{E_2}{E_1}\cos(\theta-\alpha)\right), \quad (6)$$

where r_0 is the classical electron radius , K is the magnitude of the scattering vector , P_c and P_l are the degree of circular and linear polarization of the beam , respectively , and the quantity σ is ± 1 depending on the direction of the magnetic field . The α is defined as the angle between the direction of the incident X-ray and the scattering vector . [6][7]

2.2 Experimental details

The single crystals of USe were grown by a Bridgeman method at the Oarai Branch of Institute of Material Research , Tohoku University . The specimen of USe was fixed on a Cu sample holder with radius of 12mm and was sealed by a Kapton foil with thickness of 12.5μm under 1 atomosphere of He gas to avoid the leakage of nuclear fuel substance in environments . The sample holder was then mounted on the top of the cold finger of the cryogenic refrigerator . The magnetic Compton scattering experiment was carried out at the NE-1 beam line of Accumulation Ring in National Laboratory for High Energy Physics (KEK) . The experimental setup of this experiment is shown in Fig.1 . The incident X-rays emitted from the elliptical multipole wiggler and a doubly bent by Si(111) monochromator[8] were impinged onto the sample and the scattered X-rays were detected with 13-segmented solid state detectors (SSD) of germanium located about 90cm apart from the sample . The average scattering angle was 160° ± 2° . The magnetic field of 5kOe generated by an electromagnet was applied parallel or antiparallel to the direction of the scattering vector . It should be noticed that the $< 100 >$ direction of the sample makes angle of $\phi = 15°$ from the scattering plane as shown in Fig.1(b) . The magnetic field was reversed in the sequence of $(+,-,-,+)$ with 15 second dwelling time where $+(-)$ indicates the parallel(antiparallel) direction of the magnetization relative to the scattering vector . The energy of elliptically polarized (the degree of circular polarization $P_c = 0.6$) incident beam was chosen to be 59.38 keV which is below the K edge of uranium atom . The sample was cooled by a cryogenic refrigerator to 150K (T_c of this USe is 174K) . The magnetic Compton profile , $J_{mag}(P_z)$, which is derived from the difference in cross sections when spins are reversed ($\uparrow$ to $\downarrow$) , is given by

$$J_{mag}(P_z) = \int\int \left\{ n_\uparrow(\vec{P}) - n_\downarrow(\vec{P}) \right\} dP_x dP_y, \quad (7)$$

where $n_\uparrow(\vec{P})$ and $n_\downarrow(\vec{P})$ are the momentum density of majority and minority , respectively . The $J_{mag}(P_z)$ is then normalized so as to give the spin moment μ_s as follows ,

$$\mu_S = \int J_{mag}(P_z) dP_z. \quad (8)$$

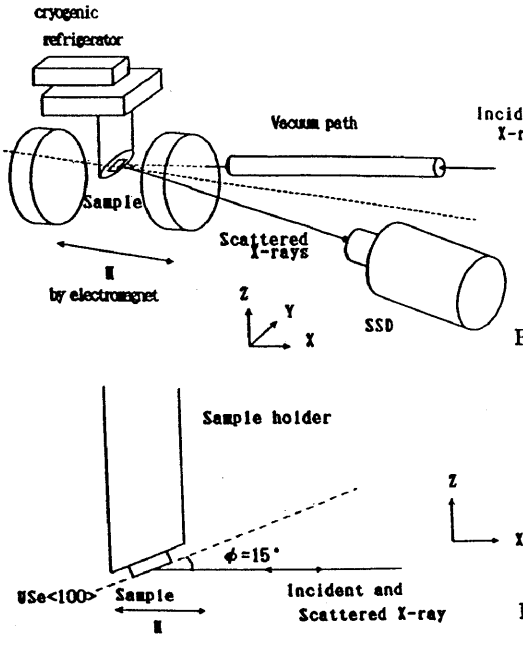

Fig1.(a)

Fig1.(b)

Figure 1: The geometry of the magnetic compton scattering experiment .

3 Results and Discussion

The observed spectra of magnetic Compton scattering , I_+-I_- , of USe and a reference sample of Fe , are shown in Fig.2 as a function of scattered X-ray energy (channel of SSD) . I_+ and I_- are intensity profiles of the Compton scattering when the magmetic field was applied parallel

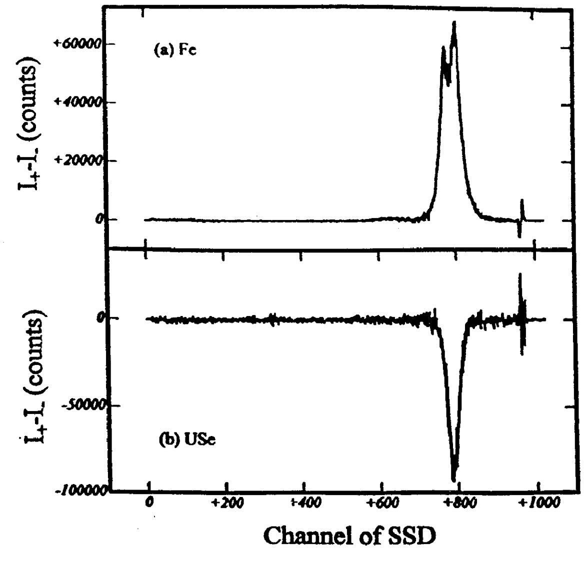

Figure 2: The spectra of magnetic Compton scattering of Fe and USe , measured at room temperature and 150K , respectively.

and antiparallel to the scattering vector direction , respectively . Fig.2 shows that the sign of I_+-I_- of USe is opposite to the sign of Fe . Fig.2 ,therefore ,shows that the spin moment of USe is antiparallel to the magnetic field in contrast to that the spin moment of Fe , i.e. 3d , is parallel to the field . This fact has been also already reported in the case of UTe[10] .

Firstly , we try to evaluate the value of the spin moment of USe . The flipping ratio R is defined as follows ,

$$R = \frac{S_+ - S_-}{S_+ + S_-}, \qquad (9)$$

where S_+ and S_- are the integrated intensity of I_+ and I_- , respectively . The whole spin moment of USe , μ_S(USe) , is given by the following equation ,

$$\mu_S(USe) = \mu_S(Fe)\left(\frac{A_{USe}}{A_{Fe}}\right)\left(\frac{R_{USe}}{R_{Fe}}\right). \qquad (10)$$

Here , R and A are the flipping ratio and the number of electrons which take part in the Compton scattering event , respectively . We use $R_{Fe} = 1.08 \times 10^{-2}$ and $R_{USe} = 8.28 \times 10^{-4}$ from the present experiment . And we also adopt $A_{Fe} = 26$ and $A_{USe} = 126 - 2$ in this experiment . The -2 come from the fact that the incident X-ray energy of the present experiment is lower than the K-edge of uranium (59.38keV) , so that 1s-electrons of U can not take part in the present Compton scattering process . The spin moment of Fe , $\mu_S(Fe)$, is 2.219 (μ_B) from the literature .

Two further corrections have to be made to estimate the value of $\mu_S(USe)$ along the direction of $< 111 >$. One comes from the fact that the magnetic field applied makes $\phi = 15°$ to the $< 100 >$ direction as mentioned before . The ratio of saturation moment along $< 111 >$ to that along $< 100 >$ is $\sqrt{3}$, therefore we have to multiply a factor $F_g = \frac{\sqrt{3}}{\cos 15°}$, to obtain $\mu_s(USe)$ along the $< 111 >$ direction . The other comes from the reduction of saturated moment because this measurement are carried out at T=150K . So we have to multiply another factor , $F_T = 1.28$. After making these two corrections , we have $\mu_s(USe) = -1.85 \pm 0.04(\mu_B)$ as the saturated value of USe spin moment along the $< 111 >$ direction . Combining this value with the whole moment , $\mu(USe) = 1.77(\mu_B)$ from the magnetization measurement , we obtain the value of orbital moment , $\mu_L(USe)$ as follows ,

$$\begin{aligned} &\mu_L(USe) \\ &= \mu(USe) - \mu_S(USe) \\ &= 1.77 - (-1.85 \pm 0.04) \\ &= 3.62 \pm 0.04. \end{aligned} \qquad (11)$$

Next , we try to decompose the MCP of USe into 5f component and the other component (conductive component) . After making the energy-dependent corrections for absorption and cross-section in momentum space , MCP of USe is shown in Fig.3 as open circle . The MCP of USe can be best fitted with calcurated MCP of uranium 5f and 6d relativistic Dirac-Hatree-Fock wave functions by the least squares method as shown in Fig.3 . The difference $J_{mag}(exp) - J_{mag}(5f)$ is shown in Fig.3 as the cross(×) and is found to be well fitted with 6d wave function . The spin moment value of 5f and 6d electrons are

$$\mu_S(5f) = -1.50 \pm 0.03(\mu_B), \qquad (12)$$

and

$$\mu_S(cond) \approx \mu_S(6d) = -0.35 \pm 0.01(\mu_B), \qquad (13)$$

from the area under each profile . The negative sign of these values show that both the spin moment of 5f electron and conductive s,p,d electrons have opposite magnetization to the bulk magnetization , which is consistent with the spin polarization measurement of the photoelectrons[9] . The similar measurement of MCP has been observed on UTe[10] .

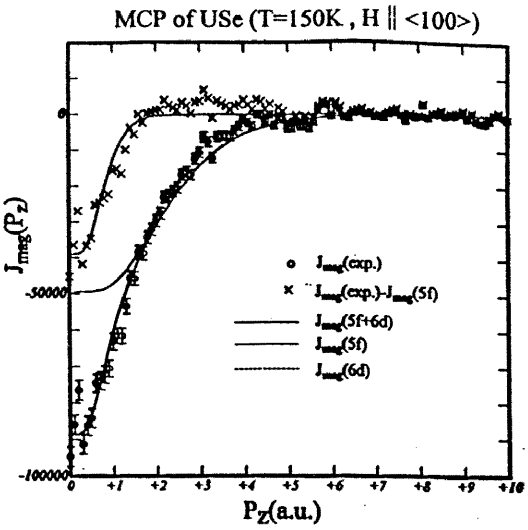

Figure 3: The magnetic Compton profile of USe . The unit of longitudinal axis is arbitrary .

If we assume that the orbital moment of USe is constructed by only 5f electrons of uranium , that is to say , $\mu_L(USe) = \mu_L(5f)$, then the total magnetic moment of 5f electrons of USe is obtained as

$$\begin{aligned} &\mu(5f) \\ &= \mu_L(5f) + \mu_S(5f) \\ &= (3.62 \pm 0.04) + (-1.50 \pm 0.03) \\ &= 2.12 \pm 0.05(\mu_B). \end{aligned} \qquad (14)$$

These values are summarized in Table 1 . From Table 1. , it is seen that

1. The 5f moment , $\mu(5f)$, are consistent with the results of neutron scattering experiment ,
2. The conductive spin moments of USe , $-0.35\mu_B$, is smaller than the value $-0.23\mu_B$, which has been suggested by a combination of neutron scattering experiment[11] and magnetization measurement .

Now , let us estimate the orbital moment and spin moment based on Hund's coupling model . Using Landé g factor and the total angular momentum number J , these moments are expressed as follows ,

$$\mu_S = (2g - 2)J \tag{15}$$

and

$$\mu_L = (2 - g)J. \tag{16}$$

Table 2 shows the calcurated values for 5f electron of U based on Hund's model . The comparison of Table 1 and Table 2 indicates that each moments differ from the calcurated value assuming Hund's coupling schemes on the case of USe .

4 Conclusion

The magnetic Compton profile (MCP) of USe was observed at 150K . The sign of profile shows that the spin moment of USe is opposite to the bulk magnetization . Combining with the value of bulk magnetization , the value of orbital magnetization was deduced , which is the same sign as bulk magnetization . Futhermore , decomposing MCP of USe into theoretical 5f and 6d contribution , it is shown that both 5f spin moment and conduction spin moment are anti-parallel to the bulk magnetization . The calcuration of Hund's coupling model implies that the simple Hund's coupling model does not completely describe the electronic structure of 5f state .

5 Acknowledgments

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Table 1: Magnetic moments obtained from the present MCP experiment and neutron scattering experiment[11] and magnetization measurement .

	$\mu(All)$	$\mu_S(All)$	$\mu_L(5f)$	$\mu_S(5f)$	$\mu_S(cond)$	$\mu(5f)$	$\frac{\mu_L(5f)}{\mu_S(5f)}$
MCP	1.77	-1.85±0.04	3.62±0.04	-1.50±0.03	-0.35±0.01	2.12±0.05	-2.41
Neutron	1.77	-	-	-	-0.23±0.1	2.0±0.1	-

Table 2: Calculated values of 5f electron of uranium based on Hund's model .

	L	S	J	g_J	$\mu_L(5f)$	$\mu_S(5f)$	$\mu(5f)$	$\frac{\mu_L}{\mu_S}$
$f^1(U^{+5})$	3	$\frac{1}{2}$	$\frac{5}{2}$	$\frac{6}{7}$	2.86	-0.71	2.15	-4.03
$f^2(U^{+4})$	5	1	4	$\frac{4}{5}$	4.80	-1.60	3.20	-3.00
$f^3(U^{+3})$	6	$\frac{3}{2}$	$\frac{9}{2}$	$\frac{8}{11}$	5.73	-2.45	3.28	-2.34
$f^4(U^{+2})$	6	2	4	$\frac{3}{5}$	5.60	-3.20	2.40	-1.75