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LETTER TO THE EDITOR

The magnetic Compton profile of UTe

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Abstract. We have performed a magnetic Compton scattering experiment with a ferromagnetic actinide compound of UTe. We have combined the results of the magnetic Compton scattering experiment and the magnetization measurement and have deduced the spin and the orbital magnetic moments. Furthermore, we have decomposed the spin moment into a 5f component and a conduction-electron-like component. We have obtained the spin moment of the 5f electron $\mu_S(5f) = -1.21\mu_B$, the orbital moment of the 5f electron $\mu_L(5f) = 3.48\mu_B$ and the spin moment of the conduction-electron-like component $\mu_S(s, p, d) = 0.36\mu_B$.

Correlated electron systems have a wide range of f electron delocalization varying from 'almost localized' to 'almost itinerant' in nature. Techniques to calculate hybridization and exchange Coulomb interaction in the f electron systems have been developed from the 'core point of view'. Such a procedure is appropriate for almost localized systems, i.e. typically cerium systems where the electrons of interest are 4f. However, 5f electron states in uranium compounds are much more delocalized than 4f electron states in rare-earth compounds because f charge leakage from a muffin-tin sphere is larger than that of rare-earth 4f systems. Thus the mutual interaction between hybridization and f self-banding is strong in uranium compounds, and the core point of view fails to reflect much of the reality [1].

Uranium monotelluride (UTe) has been studied as an 'almost itinerant' 5f compound. It has a NaCl-type crystal structure and has a ferromagnetic ordering temperature of $T_c = 104$ K [2]. It is also known to be a high-T_c dense Kondo material [3]. The magnetic moments are strictly confined to the $\langle 111 \rangle$ easy axis up to 200 kOe [2]. Magnetization and magnetic neutron scattering studies [4] give a value for the 5f ordered moments of $2.25\mu_B$ and estimate conductive s, p, d moments of $-0.34\mu_B$. The recent explicit orbital-polarized band structure calculation [5] underestimates the total magnetic moment, and the combination of two-ion interaction and LMTO band structure calculations [1] overestimates the total magnetic moment. The cause of the discrepancies between the calculations and the experiments is related to how the calculation compromises between the two extremes of localization and itineracy [1]. Presumably localized character significantly affects the orbital magnetization, and the essential physics of the magnetism of the actinide system requires the determination of not only the total moments but also the spin moments and the orbital moments individually.

Recently, it has been reported that a newly developed magnetic Compton scattering experiment reflects only spins [6, 7], and therefore the spin contribution μ_S and the orbital contribution μ_L of the magnetization are separated by combining magnetic Compton scattering experiments with magnetization experiments, because the latter reflects the total magnetization $\mu = \mu_S + \mu_L$. This procedure has been successfully applied to $HoFe_2$ and the orbital moments μ_L are discussed as a function of temperature [8]. As a spin–orbital separation method, the magnetic Compton scattering experiment has the advantage of giving the values of μ_S and μ_L, whereas in magnetic neutron diffraction analysis there is some ambiguity in the fitting parameters used to determine μ_S and μ_L. Preliminary results of the magnetic Compton measurements on UTe have been already reported in [9]. The magnetic form factor measurement of UTe by x-ray magnetic diffraction will be published in [10].

The magnetic Compton experiment yields a one-dimensional projection of spin density in momentum space, $J_{mag}(P_z)$, defined below:

$$J_{mag}(P_z) = \int \int \{n^{\uparrow}(P) - n_{\downarrow}(P)\} \, dP_x \, dP_y. \tag{1}$$

Here, $n(P)$ is the momentum density, and ↑ and ↓ denote spin-up and spin-down states, respectively. $J_{mag}(P_z)$ is called the magnetic Compton profile (MCP). The MCP gives individual information on the magnetic electrons such as conductive s, p, d electrons and 4f and/or 5f electrons, because each electron has a characteristic momentum distribution. The main purpose of this study is to measure the spin moment of 5f electrons and conductive s, p, d electrons.

The magnetic Compton scattering measurements were carried out at the beam line NE1 of the Accumulation Ring at KEK. Elliptically polarized synchrotron radiation with a degree of circular polarization of about 0.6 was produced by an elliptical multipole wiggler [11, 12]. The incident x-ray energy was chosen to be 59.38 keV which is below the U K edge. The x-rays were monochromatized with a resolution of $\Delta E/E \leqslant 10^{-3}$ and focused with a newly developed water-cooled doubly-bent Si(111) monochromator [13]. The incident beam, of approximate flux 10^{12} photons s^{-1} mm^{-2} at the sample position, was adjusted to a size of vertical height 1.3 mm and of horizontal width 3 mm.

A single-crystal sample of UTe was made by the Bridgman method at the Oarai Branch of IMR of Tohoku University. The sample was cleaved along the (100) plane and shaped into a rectangle 8 mm × 5 mm with 1 mm thickness, and mounted in a sample holder. The holder was sealed by polyimide film, with a thickness of 12.5 μm, to avoid sample oxidization and environmental contamination. The holder was set on a cryogenic refrigerator, and a magnetic field of 5 kOe was applied along the ⟨100⟩ direction by an electromagnet. The angle between the incident beam and the magnetization of the sample was set to 15°.

The scattered radiation was detected with an integrated set of 13 intrinsic germanium detectors, but only 10 detector segments were used to acquire data at the time of measurements. The detector set was located about 90 cm apart from the sample. The average scattering angle was 160° ± 2°, which depended upon the position of each detector in the horizontal scattering plane. The counting rate for the detector was adjusted to be below 25 000 cps at an initial ring current of 25–30 mA. The resolution of each detector was deduced from the width of the elastic peak, which was 0.75 au in the full width at half maximum in momentum space.

The spectra were collected in the sequence $+ - - +$ with a 15 s measurement time for each + and − and a 2 s interval between successive measurements; + and − indicate parallel and anti-parallel alignment of the scattering vector of the photon and the magnetization field, respectively. We shall refer to + and − as 'spin up' and 'spin down', respectively. The temperature of the sample was set at 80 K below the sample Curie temperature in order to saturate the magnetization with 5 kOe [9].

Figure 1 shows the spectra of magnetic Compton-scattered x-rays, $I_+ - I_-$, of Fe and UTe. Here, I_+ and I_- are intensity profiles of the Compton-scattered x-rays from the spin-up (+) and the spin-down (−) states, respectively. It must be noticed that the sign of $I_+ - I_-$ is positive for Fe, while it is negative for UTe. The negative sign of $I_+ - I_-$ is the first observation in MCP study.

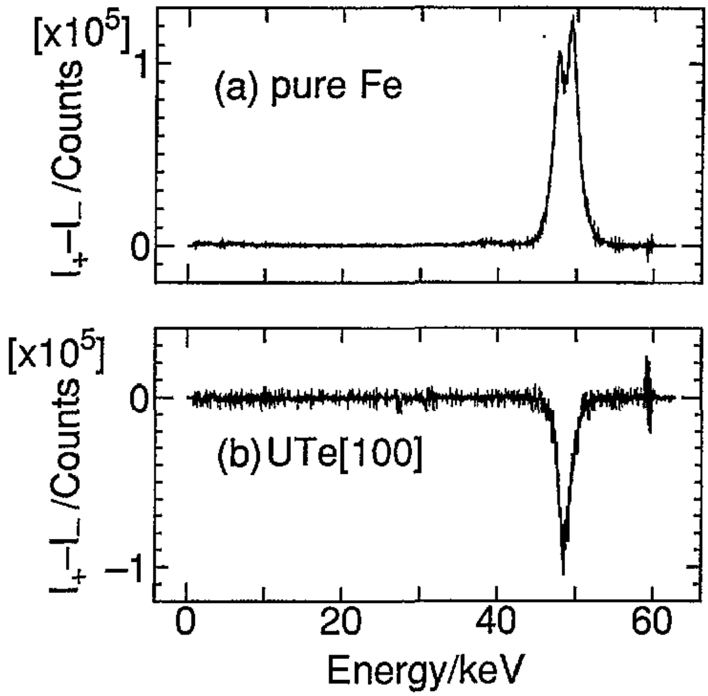

Figure 1. The spectra of magnetic Compton-scattered x-rays, $I_+ - I_-$, of (a) polycrystalline Fe and (b) single-crystal UTe. Here, I_+ and I_- are intensity profiles of Compton-scattered x-rays for spin up (+) and spin down (−), respectively.

The flipping ratio R is defined by the following equation:

$$R = (S_+ - S_-)/(S_+ + S_-). \qquad (2)$$

Here, S_+ and S_- are the integrated intensity of I_+ and I_-, respectively. The net spin magnetization of the UTe, μ_S(UTe), is given by the following equation [14]:

$$\mu_S(\mathrm{UTe}) = \mu_S(\mathrm{Fe})(A_{UTe}/A_{Fe})(R_{UTe}/R_{Fe}). \qquad (3)$$

Here, R_{Fe} and R_{UTe} are the flipping ratios of Fe and UTe, respectively. We obtained $R_{Fe} = 1.06 \times 10^{-2}$ and $R_{UTe} = 9.33 \times 10^{-4}$. The value A is the total number of electrons per chemical formula, and A_{Fe} is given as 26 for Fe. In the case of UTe, the number of electrons, A_{UTe}, is a sum of $92 - 2$ from U and 52 from Te, i.e. 142. The subtraction of 2 for U is to account for the fact that the incident x-ray energy is lower than the U K edge, and therefore the K-shell electrons in U cannot participate in the Compton scattering. The spin magnetization of Fe, μ_S(Fe), is given as $2.2\mu_B$.

Then, two corrections should be made. One is for the anisotropy effect. The UTe has strong magnetic anisotropy; the easy axis is the $\langle 111 \rangle$ direction [1]. But μ_S in this experiment is the projection of the spin moment along the direction $\langle 100 \rangle$. So a factor of $\sqrt{3}$ should be multiplied by μ_S(UTe). The other is the reduction of the saturation moment because the measurements were carried out at 80 K. From the magnetization experiment [9], the reduction factor is 1.12. After these corrections, we have finally got the spin magnetization of $\mu_S(\mathrm{UTe}) = 1.57\mu_B$. Together with the magnetization measurements ($\mu(\mathrm{UTe}) = \mu_S(\mathrm{UTe}) + \mu_L(\mathrm{UTe}) = 1.91\mu_B$), we have got the orbital magnetization $\mu_L(\mathrm{UTe}) = 3.48\mu_B$.

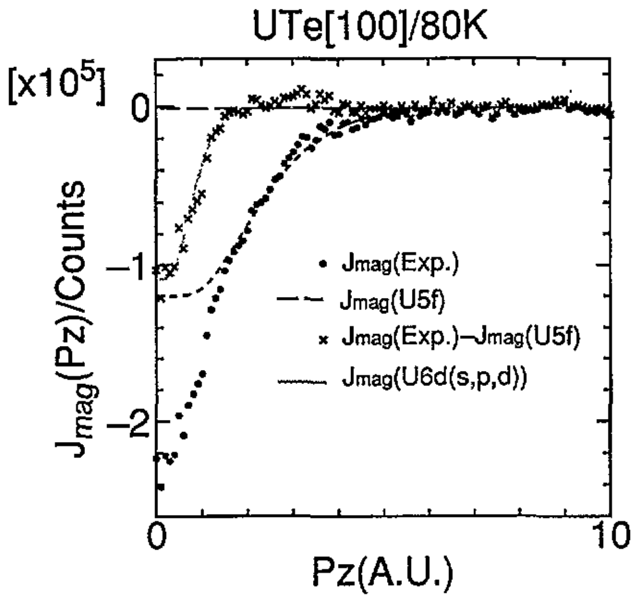

Figure 2. A comparison of the magnetic Compton profile of UTe obtained from experiment, J_{mag}(Exp), and that from calculation from U 5f wavefunctions, J_{mag}(U 5f). The difference J_{mag}(Exp) − J_{mag}(U 5f) is also compared with the profile J_{mag}(U 6d) calculated from the U 6d wavefunction.

Figure 2 shows the MCP of UTe, $J_{mag}(P_z)$, after making the corrections for absorption and cross section [15] in momentum space. It must be emphasized that the MCP of UTe is negative. This is direct evidence that the spin moment of UTe is aligned anti-parallel to the magnetic field, which means in turn that the orbital moment is aligned parallel to the magnetic field.

To see the nature of the spin moment, we have tried to decompose the MCP into 5f and conductive s, p, d characters. The wavefunctions of the U 5f electron in a free atom from Biggs *et al* [16] were used as the trial MCP for the 5f character. The theoretical profile reproduces the experimental profile very well in the momentum range above $P_z = 2.0$ au. The difference between the experimental MCP and the theoretical 5f profile is also shown as a dotted curve in figure 2. The low-momentum components of this difference profile are found to be represented very well by the wavefunction of U 6d electrons in a free atom, which is more 'delocalized' than that for the U 5f electrons.

The low-momentum components ($P_z < 2$ au) are ascribed to spin-polarized conductive s, p, d electrons, which are induced by RKKY interactions with 5f electrons. From the above fitting analysis of the MCP, the 5f components of UTe have a value of $-1.21\mu_B$ and the conductive s, p, d components have a value of $-0.36\mu_B$. This means that both the spin components of 5f and conductive s, p, d electrons have negative polarization. The negative spin polarization of conductive s, p, d electrons, which is dominated by U 6d electrons, is consistent with spin-polarized photoemission experiments by Erbudak and Meier [17]. However, they guessed that the 5f spin magnetization couples in an anti-parallel fashion to the conductive s, p, d spin magnetization as compared with their band calculation [18]. Recently calculations by Brooks *et al* have shown that the 5f spin is always parallel to the 6d spin, due to both 6d–5f hybridization and local exchange interactions [19]. The present results support the theoretical calculations by Brooks *et al*.

Here, we assume that the main orbital contribution to the magnetization comes from 5f electrons, say $\mu_L(5f) = \mu_L(\mathrm{UTe})$. The total magnetic moment of 5f electrons of UTe is then given as

$$\mu(5f) = \mu_L(5f) + \mu_S(5f) = 3.48 - 1.21 = 2.27\mu_B. \tag{4}$$

The values of each of the components of the magnetization are summarized in table 1. The results of the present MCP are in fairly good agreement with those of the magnetic neutron diffraction experiment [3].

Table 1. Magnetic moments of UTe compound, in units of Bohr magnetons.

	μ_S(UTe)	μ(5f)	μ_S(s, p, d)	μ_S(5f)	μ_L(5f)	$\mu_L(5f)/\mu_S(5f)$
UTe(MCP)	1.91[3]	2.27	−0.36	−1.21	3.48	2.88
UTe(Neutron)[3]	1.91	2.25	−0.34	?	?	?
f^2(Hund)	—	3.20	—	−1.60	4.80	3.00
f^3(Hund)	—	3.27	—	−2.46	5.74	2.33

According to Hund's coupling scheme of a free ion of f electron systems, the total angular momentum number J is a good quantum number, and the spin contribution and orbital contribution of the magnetization (μ_S and μ_L) are given as follows:

$$\mu_S = (2g-2)J \qquad \mu_L = (2-g)J. \tag{5}$$

Here, g is the Landé g-factor. The calculated μ_S, μ_L and the ratio $-\mu_L/\mu_S$ for the f^2 configuration (U^{4+}) and f^3 configuration (U^{3+}) are shown in table 1. In both configurations, however, the magnetic moments of the spin and the orbital contribution in UTe are overestimated. Furthermore, crystal field effects under cubic symmetry are taken into account and the orbital magnetization and spin magnetization are estimated. However, no states under the crystal field can reproduce the experimental results in both the f^3 configuration and the f^4 configuration. The results obtained from calculation suggest that the localized picture of the 5f electron is invalid in the UTe. The dense Kondo behaviour comes from the strong hybridization between the 5f electrons and the conduction electrons, and recent inelastic neutron experiments on the magnetic excitation suggested that 5f electrons were strongly hybridized with the conduction electrons [20]. Then we suggest that the 5f electrons are rather itinerant and the orbital magnetization is partially quenched due to the strong 5f-conduction hybridization.

In conclusion, magnetic Compton scattering experiments of UTe have been carried out at 80 K. It was found that the spin magnetization μ_S couples anti-parallel to the orbital magnetization μ_L, and the orbital magnetization is parallel to the bulk magnetization. The spin of 5f electrons couples parallel to one of the s, p, d conductive electrons. The obtained value of the spin magnetization and the orbital magnetization cannot be explained by the localized picture. Theoretical analysis including correct spin-orbit interaction may be necessary for precise shape analysis of the MCP in UTe in order to clarify the magnetic behaviour of the 5f electrons.

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