



	Experiment title: Study of the high pressure phases of the triplet superconductor UTe ₂	Experiment number: HC4373
Beamline:	Date of experiment: from: 16/06/2021 to: 21/06/2021	Date of report: 12/09/2021
Shifts:	Local contact(s): A. Rogalev	<i>Received at ESRF:</i>
Names and affiliations of applicants (* indicates experimentalists): D. Braithwaite* CEA- Grenoble G. Knebel CEA – Grenoble J.P. Sanchez* CEA- Grenoble F. Wilhelm* ESRF- Grenoble		

Report:

The aim of the beamtime was to study with XMCD the high pressure phases of the triplet superconductor UTe₂. First we have investigated the magnetism of UTe₂ at ambient pressure by performing XMCD, and in a second time, we have performed high pressure XANES measurements at 2.7 K. Due to a technical problem with the 8 T superconducting magnet, we could not investigate the magnetic part under pressure as described in the proposal.

1) XANES and XMCD at the U- M_{4,5} edges at ambient pressure

The XANES and XMCD spectra recorded at 2.7 K in an applied field of 17 T are shown in Fig.1. The branching ratio determined as $B = I_{M5} / (I_{M5} + I_{M4})$ where I_{M5} and I_{M4} are the integrated areas of the white lines at the M₅ and M₄ edges, respectively is found to be 0.706(2). Its value lies between those of the U³⁺ (0.729) and U⁴⁺ U free ion values. Thus, the U ions are in an intermediate valence state as expected from band structure calculations where the 5f count was found to be close to 2.8. Taking this value and using the spin orbit sum rule, we may evaluate the number of electrons in the individual shells corresponding to $j = 7/2$ and $j = 5/2$, i.e., 0.28 and 2.52, respectively.

The XMCD spectrum was recorded at 17 T with H applied along the **a** easy axis. A large signal has been detected at both M_{5,4}-edges under 17T.

Using the orbital magneto-optical sum rule, we estimated the 5f orbital (μ_L) and spin (μ_S) moments assuming that the 5f count is equal to 2.8, 3 and 2 and taking theoretical values for the $\langle T_Z \rangle / \langle S_Z \rangle$ ratios ($\langle S_Z \rangle$ and $\langle T_Z \rangle$ are the z- components of the spin and magnetic dipole operators, respectively). It was concluded that the 5f² (U⁴⁺) configuration led to unphysical results and that the actual 5f count is close to 2.8.

The final results of our analysis are : $\mu_L = 1.61 \mu_B$, $\mu_S = - 0.76 \mu_B$ and $-\mu_L / \mu_S = 2.12$. The orbital to spin moment ratio falling below the free ion U³⁺ value (2.60) indicates significant delocalization of the 5f electrons due to the hybridization with the Te *spd* bands. Furthermore it was shown, by recording the maximum of the XMCD signal at the M₄ edge, that the U magnetization matches well with the macroscopic magnetization.

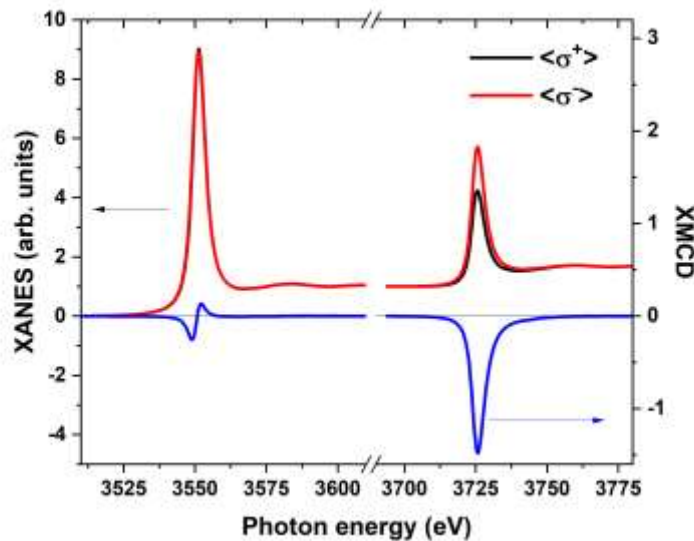


Fig.1 XANES and XMCD spectra at the U- $M_{4,5}$ edges for UTe_2 at 2.7 K and applied along the easy axis **a**.

2) XANES and XMCD at the Te- $L_{1,2,3}$ edges at ambient pressure

The XMCD signal at the Te- L_1 edge is only due to the orbital polarization of the $5p$ Te unoccupied states through hybridization with the U spin-orbit split states. The integration of the negative and positive peaks led to an overall negative signal which indicates a positive $5p$ orbital moment whose magnitude is difficult to estimate. No information is provided for the $5p$ spin moment.

The XMCD signal at the Te- $L_{2,3}$ edges is minute because the small number of holes inside the $4d$ band.

The total $4d$ moment is tentatively estimated to amount about $+0.001\mu_B$.

To conclude the Te $5p$ moment should be the main contribution to the Te moment, in agreement with ARPES and bnd structure calculations of the partial densities of states.

3) High pressure XANES at the U- $M_{4,5}$ edges

So far the high pressure (HP) data were limited to XANES. The 8 T superconducting cryomagnet forseen for the XMCD experiments was sent for repair to Oxford Instruments.

Fig.2 shows selected data for the pressure dependence of the M_5 white line. It indicates that the number of holes in the $5f$ shell first increases while it decreases at the highest pressure. Thus, the U valence first increases towards U^{4+} then decreases.

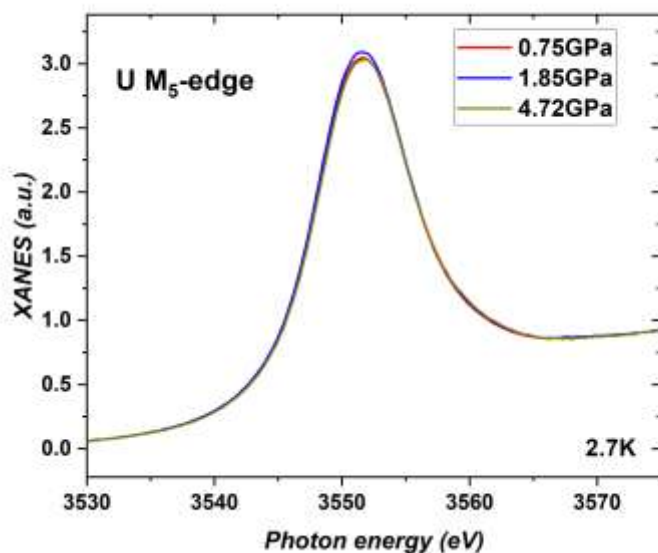


Fig.2. Pressure dependence of the U- M_5 white line for UTe_2 recorded at 2.7 K