ESRF	**Experiment title:** Magnetism of Neptunium in NpX_2 (X=Al, Fe, Os) ferromagnetic compounds probed with XMCD	**Experiment number**: HE-2561
Beamline: ID12	**Date of experiment**: from: 30/04/2008 to: 05/05/2008	**Date of report**: 24/09/2008
Shifts: 18	**Local contact(s)**: Andrei Rogalev	*Received at ESRF:*

Names and affiliations of applicants (* indicates experimentalists):

Dr. Fabrice WILHELM*, *ESRF*
Prof. Roberto G.M. CACIUFFO*, *ITU Karlsruhe, Germany*
Dr. Rachel ELOIRDI, *ITU Karlsruhe, Germany*
Dr. Eric COLINEAU, *ITU Karlsruhe, Germany*
Prof. Gerard LANDER*, *ILL*
Dr. Andrei ROGALEV*, *ESRF*
Dr. Ross SPRINGELL*, *ESRF*
Prof. William G. STIRLING*, *ESRF*

Report:

In this experiment we have examined three different Np compounds, all with the same cubic *fcc* Laves crystal structure (C15), namely $NpAl_2$, $NpFe_2$ and $NpOs_2$. All are ferromagnetic with a Curie temperature of 56K, >300K, 10K respectively and have a magnetic moment of 1.22 μ_B/f.u., 3.20 μ_B/f.u., 0.60 μ_B/f.u. These materials have been chosen because $NpAl_2$ is considered a localised system, whereas $NpOs_2$ is itinerant. In $NpFe_2$ there is evidence of strong hybridisation between Np and Fe atoms. All samples have been prepared at ITU, Karlsruhe, and have been carefully characterized structurally and magnetically by means of X-ray Diffraction and SQUID magnetometry. The macroscopic magnetic properties of the sample measured are in excellent agreement to those published earlier [see. Aldred et al., PRB 10 (1974) 1011; Aldred et al., PRB 14 (1976) 1276; Aldred et al., PRB 11 (1975) 530]. All Np based compounds have been encapsulated in especially designed sample holder of Cu covered with beryllium and Kapton windows. All safety procedures were respected and no problems were encountered during the setup and measurements.

As written in our proposal, we have successfully carried out all the proposed measurements. For the first time, an XMCD signal at the Np $M_{4,5}$-edge in ferromagnetic $NpAl_2$, $NpFe_2$ and $NpOs_2$ have been measured. As expected, the observed dichroic signal is larger at the *M*4 compared to the *M*5 absorption edge but to a lesser extent than in U based ferromagnets. Fig.1 and Fig.2 show the XAS and the corresponding XMCD signal recorded at the Np *M5* and *M4*-edges in $NpFe_2$ and $NpOs_2$. We observe a strong dichroic signal with a similar spectral shape for the two compounds. The site and orbital selectivity, typical of absorption spectroscopy, together with the existence of sum rules, will allow us to calculate the expectation value of the Np 5*f* orbital magnetic moment of the ground state from the XMCD experimental spectra. Even the expectation value of the Np 5*f* spin magnetic moment of the ground state can be evaluated, if a reasonable approximation of the dipolar moment can be provided. The study of the branching ratio can therefore bring valuable information regarding the coupling scheme which best describes the Np electronic and magnetic properties, within which to discuss the actinides and therefore implies a different weighting of the dipolar

contribution compared to the spin one. Work is in progress regarding the evaluation of the different magnetic moment contributions.

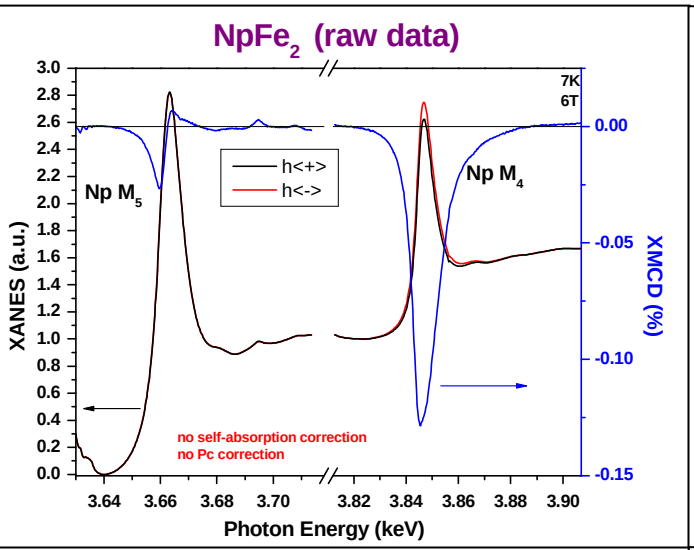

Fig.1: Normalized XAS (left scale) and XMCD (right scale, blue line) recorded at the Np $M_{5,4}$-edges in $NpFe_2$ compounds under 6Tesla and at 7K. Raw data are shown, without correction for self-absorption effects and for incomplete circular polarization rate.

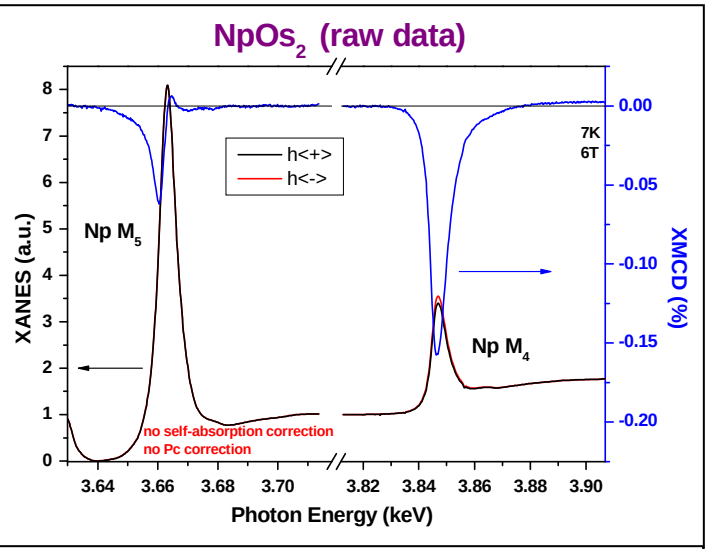

Fig.2: Normalized XAS (left scale) and XMCD (right scale, blue line) recorded at the Np $M_{5,4}$-edges in $NpOs_2$ compounds under 6Tesla and at 7K. Raw data are shown, without correction for self-absorption effects and for incomplete circular polarization rate.

We have also measured an XMCD signal at the Fe *K*-edge in $NpFe_2$ (Fig.3) that will be compared to the XMCD signal recorded in UFe_2 (same Laves phase structure). We have observed a large influence of the strong Np spin-orbit interaction on the XMCD signal that is essentially located in the pre-edge peak. Finally, a sizeable XMCD signal up to 2% with respect to the edge jump at the Os $L_{2,3}$-edges has been measured in $NpOs_2$ (Fig.4) implying the existence of a finite Os 5*d* induced magnetic moment. Using the sum rules, it would be possible to determine individually the spin and orbital magnetic moment induced by Np. We can already say that the 5*d* spin moment is aligned parallel to the Np total moment. A theoretical effort based on fully relativistic band structure calculations on both Np and Os sites is under progress.

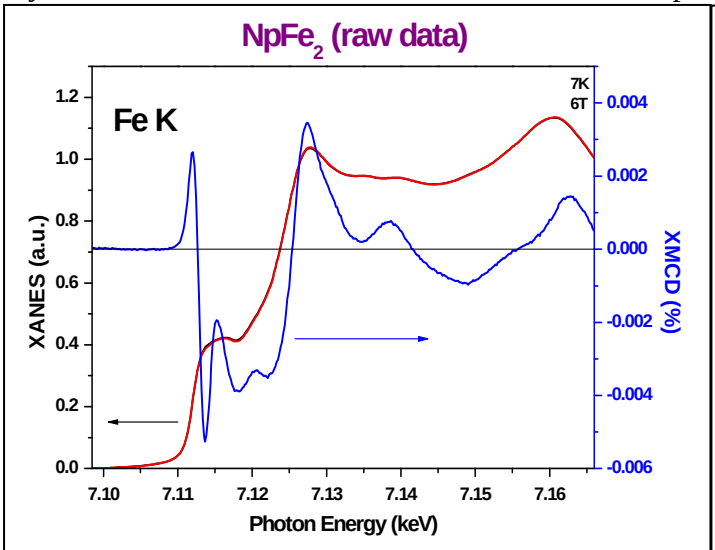

Fig.3: Normalized XAS (left scale) and XMCD (right scale, blue line) recorded at the Fe *K*-edge in $NpFe_2$ compounds under 6Tesla and at 7K. Raw data are shown, without correction for self-absorption effects and for incomplete circular polarization rate.

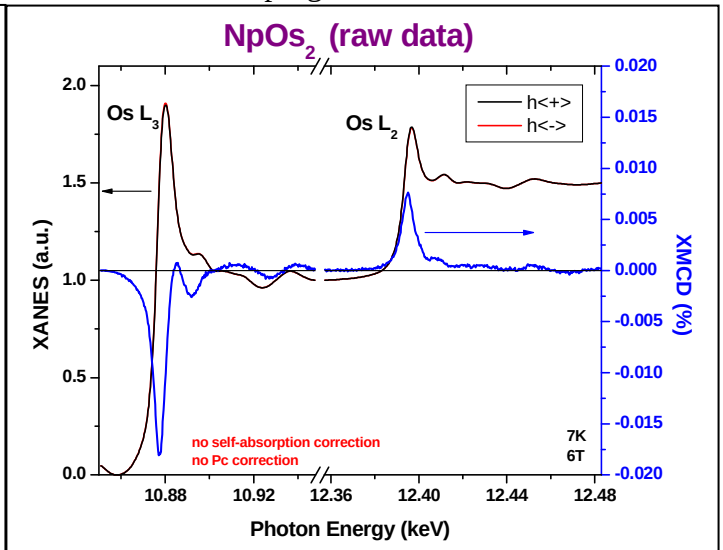

Fig.4: Normalized XAS (left scale) and XMCD (right scale, blue line) recorded at the Os $L_{3,2}$-edges in $NpOs_2$ compounds under 6Tesla and at 7K. Raw data are shown, without correction for self-absorption effects and for incomplete circular polarization rate.