μSR ON NEPTUNIUM INTERMETALLICS AND OXIDES

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We have studied UO_2, NpO_2 and $NpCo_2$. The uranium was completely depleted (~5 g), the neptunium compounds were of ^{237}Np, ~1.5 g with an activity of about 40 MBq. All samples were kept in a helium atmosphere within double containers made from ultrapure Al. The helium leak tightness of the containers was better than 10^{-9} atm cm^3 s^{-1}. The magnetic behaviour of UO_2 and NpO_2 has been studied for many years, but as yet no consistent description has emerged. The crystal structure at room temperature is of the cubic face centered fluorite-type. It is expected that the magnetic behaviour of these nearly insulating compounds may be described by a localized picture. Complications arise, however, due to the presence of magneto-elastic and quadrupolar interactions. UO_2 becomes antiferromagnetic below 30.8 K (see *e.g.* [1] and references given therein). The first order transition is accompanied by a weak lattice distortion. Neutron diffraction experiments support a non-collinear spin structure, probably of 3k-type [2]. Susceptibility, resistivity and specific heat data for NpO_2 indicate a phase transition at 25 K. Neither neutron diffraction nor Mössbauer spectroscopy (see *e.g.* [3]) have revealed the presence of ordered magnetic moments, putting an upper limit of $0.02\,\mu_B$ on them.

Our ZF μSR on UO_2 reveal a clear magnetic transition at about 30 K with the appearance of at least three spontaneous frequencies. The temperature dependence follows that of the sublattice magnetization: after a quick rise below T_N (which is typical for a first order transition) a gradual saturation is achieved at 57 MHz, 75 MHz and 103 MHz, respectively.

Also in NpO_2, two spontaneous precession frequencies (about 6 MHz and 13 MHz) develop below 25 K. The precession signals represent, however, only about 10 % of the sample. The rest of the signal can be described by an exponential. Its damping scales with the temperature dependent spontaneous frequencies. These results give the first evidence of magnetic order, though part of the moments may be frozen in a severely inhomogeneous surrounding. Comparing the observed frequencies with those found for UO_2 (ordered moment of about $1.7\,\mu_B$) and assuming the muon stopping sites to be the same, the ordered neptunium moment has to be $\mu_0 \sim 0.1\,\mu_B$, if the spin structure also is the same. This disagreement with the estimate from Mössbauer data leads to the conclusion that the nature of the transition is different in UO_2 and NpO_2.

The magnetic behaviour of the Np C15 Laves-phase compounds is strongly influenced by $5f$ to ligand (transition metal or Al) d- or p-electron hybridization. The resultant f-band broadening causes a strong reduction of the Np moments in the ordered state similar to heavy fermion compounds. Earlier μSR measurements on rare earth (RE) C15 compounds have shown that the magnetic transitions can readily be traced from the development of damping with temperature. On RE dialuminides no spontaneous precession could be observed in the ordered state [4], while RE-Fe_2 show a signal [5]. Earlier tests on $NpAl_2$ [6] also revealed the onset of spontaneous precession below $T_c = 56$ K. ^{237}Np Mössbauer spectroscopy on $NpCo_2$ shows an onset of magnetic hyperfine splitting below 13 K [7]. The ordered moment is about $0.5\,\mu_B$. The shape of the spectra, however, indicates either a wide inhomogeneous broadening due to some randomness in spin structure or the presence of rather slow dynamics even at 1.5 K. Susceptibility data point to an antiferromagnetic transition. There is, however, no evidence for antiferromagnetic order from elastic neutron scattering.

Our μSR-data on $NpCo_2$ reveal the development of at least one spontaneous frequency below 13 K. This proves the existence of magnetic order agreeing with Mössbauer measurements [7]. The precession signal is strongly damped and can only be resolved within the first $0.1\,\mu$s which corresponds to a wide inhomogeneous broadening.

References

[1] G. Amoretti *et al.*, Phys. Rev. B **40** (1989) 1856.

[2] P. Giannozzi and P. Erdös, J. Mag. Mag. Mater. **67** (1987) 75.

[3] J.M. Friedt *et al.*, Phys. Rev. B **32** (1985) 257.

[4] O. Hartmann *et al.*, Phys. F **16** (1986) 1593.

[5] S. Barth *et al.*, Phys. Rev. B **33** (1986) 430.

[6] K. Aggarwal *et al.*, Hyperfine Interact. **64** (1990) 401.

[7] A.T. Aldred *et al.*, Phys. Rev. B **11** (1975) 530.