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237Np Mössbauer effect study of NpPdSn

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During the past decades ternary actinide-based compounds containing d- and p-electron elements have attracted much attention for their large variety of physical behaviors arising mostly from the hybridization of actinide 5f electrons with s, p and /or d states on neighboring sites. An example of a system with promising physical behavior is the ATX composition where A stands for actinide atoms, T for d-element metals and X for p-electron elements.

Among this family, the uranium stannite UPdSn exhibits an antiferromagnetic phase transition at 37 K leading to a magnetic structure with orthorhombic symmetry. A second transition occurs at 25 K and the magnetic structure becomes of monoclinic symmetry [1]. The plutonium analogue PuPdSn undergoes two successive antiferromagnetic transitions at 21 K and 9.6 K [2]. In contrast, the neptunium analogue NpPdSn exhibits only one antiferromagnetic transition at 19 K [3]. An effective magnetic moment $\mu_{\text{eff}} = 2.66 \mu\text{B}$, close to the Np³⁺-free ion value, was inferred from magnetization measurements but the ordered moment was not determined in previous studies. Here, we present a ²³⁷Np Mössbauer investigation of the NpPdSn compound.

The ATX compounds crystallize in various crystal structures, like the hexagonal ZrNiAl- and GaGeLi- types, the orthorhombic TiNiSi- type and the cubic MgAgAs-type.

NpPdSn crystallizes into the hexagonal ZrNiAl-type structure with space group P-62m. A Rietveld analysis of the x-ray powder diffraction profile measured at room temperature has been performed to refine the lattice parameters and the internal atomic coordinates. The diffraction pattern is shown in Figure 1, together with the calculated profile and the residual line. The results show that the investigated sample is single-phase, with lattice parameters $a = 7.5075 \text{ \AA}$ and $c = 4.0954 \text{ \AA}$, in agreement with previous investigations [3]. The Np atoms occupy the 3f site ($x=0.5875$), the Pd atoms are located on the 1a and 2c sites and the Sn atoms fill the 3g site ($x=0.2487$).

²³⁷Np Mössbauer spectra were recorded between 4 K and 25 K in a transmission geometry spectrometer with a ²⁴¹Am source kept at a temperature of about 4 K. The absorber was prepared by grinding the sample to a fine powder to ensure a constant surface density with an optimal thickness of 140 mg Np/cm². The velocity scale was calibrated using the NpAl₂ standard compound. The isomer shift, IS, is given relative to NpAl₂. The spectra were analyzed using Lorentzian lines. The position and relative intensities of the absorption lines of Np nuclei were calculated by solving the complete Hamiltonian for the hyperfine interaction in both the excited and ground nuclear state of ²³⁷Np.

Mössbauer spectra of NpPdSn at 25 K and 4.2 K are presented on the Figure 2.

Following the value of IS = 14.4 mm/s (vs NpAl₂) we assigned a formal 3+ oxidation state to Np atoms [4], which is in agreement with the magnetization data.

In the paramagnetic region, the spectrum consists in only one a small quadrupolar splitting (QS), reflecting the fact that Np occupies a unique crystallographic site.

At 4K, the spectrum is split by the magnetic hyperfine interaction caused by the ordering of the magnetic moments. However, several subspectra - with equal IS and QS parameters but different magnetic hyperfine field B_{hf} - are required to reproduce the experimental pattern. In our fit, we used three different values of B_{hf} , corresponding to three different magnetic moments carried by the neptunium atoms. Two of the measured moments have a similar value ($\mu_{\text{Np}} = 1.53(5) \mu\text{B}$ and $1.44(5) \mu\text{B}$), the third one is almost vanishing ($\mu_{\text{Np}} = 0.05(5) \mu\text{B}$). The latter is indeed visible on the central line of the 4 K spectrum.

The higher number of subspectra than expected from the crystallographic structure may be due to different orientations of the principal axis (V_{zz}) of electric field gradient of Np atoms at 3f site vs. crystallographic axis, as observed in FeSn, crystallizing also into an hexagonal crystal structure [5]. The existence of several subspectra with different magnetic moments could also originate from a complex magnetic structure, like e.g. in NpIn₃ where a modulated antiferromagnetic order, including vanishing moments, was observed by

Mössbauer spectroscopy and neutron diffraction [6].

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