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THE PROBABILITY OF NON-RADIATIVE DECAY OF THE 3d LEVEL IN MUONIC²³⁷Np

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The X-ray spectrum of muonic ²³⁷Np has been investigated with stopped muons in a NpO₂ target, containing about 10 g of ²³⁷Np. The probability of the radiationless muonic $3d \rightarrow 1s$ transition in ²³⁷Np, (9 ± 4) %, was obtained by comparing the relative intensities of the main muonic X-ray transitions in singles and coincidence spectra. The coincidences were gated by the $2p \rightarrow 1s$ transitions.

The role of quadrupole $3d \rightarrow 1s$ radiationless transitions in nuclear excitation, leading to prompt processes like γ decay, neutron emission or fission [1-4],

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has been discussed in several papers [5-8].

There are two theoretical descriptions of the nonradiative width of the 3d level in heavy muonic atoms. A microscopical one [5] emphasizes the role of the compound nucleus mechanism, and a phenomenological one [8] describes it in terms of the isoscalar giant quadrupole resonance as an entrance channel for nuclear excitation accompanied by the radiationless $3d \rightarrow 1s$ transition.

If the phenomenological approach is correct, the probability of the radiationless $3d \rightarrow 1s$ transition,

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calculated by Teller and Weiss [8] for muonic ²³⁸U to be about 15%, should be quite similar for muonic atoms of neighbouring nuclei such as ²³²Th, ²³⁵U or ²³⁷Np. For ²³⁸U this probability is found [5] to agree with the prediction [8]. However, so far no other experimental data are available. To provide such data was one of the motivations to perform the present experiment.

Experimentally there is a systematic and rapid increase in the probability of prompt fission with the value of the fissility parameter [1,3]. Regarding prompt fission of muonic ²³⁸U, about 75% of all events are due to the $3d \rightarrow 1s$ radiationless transition [7]. The question then arises how the yield of prompt fission is shared between the $3d \rightarrow 1s$ and the $2p \rightarrow 1s$ radiationless transitions in muonic ²³⁷Np, where the total prompt fission rate per stopped μ^- is about 10 times higher than the corresponding figure for ²³⁸U. In order to clarify these points, we performed a measurement on ²³⁷Np to determine the probability of the radiationless muonic $3d \rightarrow 1s$ transition, using the same experimental method as was used to determine the non-radiative decay of the 3d level in muonic ²³⁸U [6,7].

From a comparison of X-ray singles spectra with coincidence spectra, in which the coincidence rate between the $2p \rightarrow 1s$ transition and other cascade lines is measured, the fraction of missing $3d \rightarrow 2p$ muonic X-rays is obtained, giving the total decay probability of the 3d level not populating the 2p level. Correcting this value for the radiative width of the $3d \rightarrow 1s$ transition and disregarding the very weak ($\leq 0.1\%$) $3d \rightarrow 2s$ transition, one obtains the non-radiative width of the 3d level.

The experiment was performed at the SIN cyclotron with a beam of negative muons, having a momentum of 85 MeV/c. The beam intensity, measured as the number of coincidences between the first two counters of the beam telescope, was about 7×10^5 µ⁻/s. The muons were slowed down to stop in the ²³⁷Np target by using a beryllium/polyethylene degrader. The target consisted of NpO₂, containing 9.981 g of ²³⁷Np, with a thickness of about 0.92 g/cm². To avoid pile-up, events in which two muons arrived within a time interval of 200 ns of each other were rejected.

The previous measurement [6] on 238 U has shown the necessity to take care of the unfavourable intensities of the weak $5 \rightarrow 4$ and $4 \rightarrow 3$ transitions and of the components in the $2p \rightarrow 1s$ complex as compared to the high Compton background. To match this problem the muonic ²³⁷Np X-ray spectra were registered in a large volume (28%) intrinsic Ge detector. provided with a BGO-shield $(13 \text{ cm} \times 13 \text{ cm} \times 15 \text{ cm})$ for Compton suppression. The suppression factor for the Compton background in the measurement was about five. The maximum energy registered (in 8192 channels) by the Ge counter, was about 10 MeV. Two large-volume scintillation counters, a CsF and a NaI(T1) crystal, were used to record the $2p \rightarrow 1s$ transition in the muonic ²³⁷Np cascade. In the offline analysis these detectors were set to gate an energy interval from 4 to 8 MeV, which includes the $2p \rightarrow 1s$ transitions. The muonic ²³⁷Np spectra were registered in both singles mode and in coincidence with the CsF and/or NaI(T1) counters. All three detectors were mounted around the target at 90° with respect to the u-beam direction. The data acquisition system is described in ref. [9].

In figs. 1-3 selected parts of the muonic ²³⁷Np X-rav spectrum are presented. In the coincidence spectra of the Ge counter we observed a small amount of the $2p \rightarrow 1s$ transitions, due to accidental coincidences with the counting rate in the scintillation crystals. The observed yield of the $2p \rightarrow 1s$ transition corresponds to an admixture of the singles spectrum into the coincident spectrum of about 10-15%. After applying a correction for this effect the ratio of the gated to the non-gated L X-ray complexes are normalized to unity, using the summed contents of the main components of these transitions in the two energy intervals 3070-3095 keV and 3280-3360 keV (fig. 1). This correction on the coincidence spectrum gives an uncertainty in the final result that is negligible in comparison to the quoted accuracies. The above energy intervals, chosen for the summed components of the L X-rays, are selected to minimize the influence of the background and possible systematic errors. A change of less than 0.3% occurs in the normalization, when choosing the energy interval to be 2950-3410 keV.

In fig. 2, superimposed on each other, the gated and non-gated spectra of the $4f \rightarrow 3d$ transitions are shown and normalized as just discussed (this normalization value, obtained from the ratio of the total sum of the L X-ray complex in the singles to that of the



Fig. 1. The muonic ²³⁷Np 3d \rightarrow 2p transition measured in coincidence with the 2p \rightarrow 1s transition. The solid line (histogram) represents the singles spectrum normalized to the coincidence data. The background has been subtracted. The left-hand side muonic complex represents the $3d\frac{5}{2} \rightarrow 2p\frac{3}{2}$ hyperfine complex and the right-hand side complex represents the $3d\frac{5}{2} \rightarrow 2p\frac{3}{2}$ hyperfine complex.



Fig. 2. The muonic ²³⁷Np 4f \rightarrow 3d transition measured in coincidence with the 2p \rightarrow 1s transition. The singles spectrum (solid line histogram) was normalized to the coincidence data by multiplying the content of the original spectrum with the normalization factor, obtained from the ratio of gated to non-gated spectra of the 3d \rightarrow 2p transition. The background has been subtracted (see text). The left-hand side muonic complex represents 4f $_2^2 \rightarrow$ 3d $_2^2$ hyperfine complex and the right-hand side complex represents the 4f $_2^2 \rightarrow$ 3d $_2^2$ hyperfine complex.

coincidence spectrum, was used for all the other complexes). In a similar way, fig. 3 presents both spectra for the $5g \rightarrow 4f$ transitions. The background for the different X-ray transitions has been defined by a step-function as described e.g. by Taal et al. [10]. However, the data have also been analysed with a linearly decreasing background. The difference in the results for the two cases was within the quoted errors.

The suppression of the $4f \rightarrow 3d$ and $5g \rightarrow 4f$ transitions in the muonic ²³⁷Np X-ray cascade, when measured in coincidence with the $2p \rightarrow 1s$ transition, is clearly observed. For the separate L X-ray components in ²³⁷Np the differences in yield between singles and coincidence spectra are negligibly small within the limits of the errors (fig. 1), whereas they were significantly different in the case of ²³⁸U [6]. Because of the high background of the natural γ -ray activity of ²³⁷Np, the $6 \rightarrow 5$ transition was obscured and could not be used for the analysis.

Based on the $4f \rightarrow 3d$ and $5g \rightarrow 4f$ results, the 2p level is bypassed by $(13\pm5)\%$ and $(10\pm6)\%$, respectively, or on the average by $(12\pm4)\%$. The quoted errors include inaccuracies due to background estimation. The radiative, relative width of the 3d level in muonic ²³⁷Np, is obtained from a comparison of the singles X-ray spectra of ²³⁷Np and ²⁰⁸Pb, resulting in a value of $(3\pm1)\%$ [14] and in good agreement with the calculated value for muonic ²³⁸U [6]. As a consequence, the observed suppression of the 4f \rightarrow 3d transition in the coincidence spectrum corresponds to a probability of $(9\pm4)\%$ for a radiationless $3d\rightarrow1s$ transition in muonic ²³⁷Np.

Due to the rather large relative errors in both measurements, the smaller value of the radiationless $3d \rightarrow 1s$ transition in muonic ²³⁷Np as compared to ²³⁸U, where it was found to be $(14\pm5)\%$ [6], might not be statistically significant. These results show that the phenomenological model predictions [8] are quite good.

As the muonic $2p \rightarrow 1s$ transition energies are almost the same in these nuclei, the reason for the difference in the ratios of prompt to delayed muon induced fission yield, namely $(28.3 \pm 0.3)\%$ and $(8.8 \pm 0.3)\%$ [11-14] for ²³⁷Np and ²³⁸U, respectively, must be explained by the difference in height of fission barriers: $E_b(^{237}Np) = 5.50$ MeV and $E_b(^{238}U) = 6.35$ MeV [11-13]. In the case of ²³⁷Np,



Fig. 3. The $5g \rightarrow 4f$ transition in muonic 237 Np, treated as mentioned in the caption of fig. 2. The left-hand side muonic complex represents the $5g_2^2 \rightarrow 4f_2^2$ hyperfine complex and the right-hand side complex represents the $5g_2^2 \rightarrow 4f_2^2$ hyperfine complex.

 $(22.0\pm0.2)\%$ of all fission can be ascribed to prompt fission, resulting in a prompt fission probability of $(2.7\pm0.3)\%$ for ²³⁷Np, when using a value of $(12.4\pm1.0)\%$ for the amount of total fission per µstop [3]. Assuming most of the radiationless $3d \rightarrow 1s$ transition probability to give rise to prompt neutron events, as in the case in ²³⁸U [7], one may conclude that the radiationless $2p \rightarrow 1s$ transition plays a much more important role for prompt fission in ²³⁷Np than is in the case in ²³⁸U. This has, however, to be verified in a separate fission–X-ray coincidence experiment.

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