

# MEASUREMENT OF THE NEUTRON TOTAL AND CAPTURE CROSS SECTIONS OF $^{237}\text{Np}$ IN THE EPITHERMAL ENERGY RANGE

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## ABSTRACT

Within the framework of a collaboration between the Institute for Reference Materials and Measurements (IRMM) and the Commissariat à l'Energie Atomique (CEA), a project has been started to measure neutron cross sections for nuclear waste transmutation purposes. We describe the measurements of the neutron total and capture cross section of  $^{237}\text{Np}$ , performed at the pulsed white neutron source GELINA of the IRMM in Geel (Belgium). We used the time of flight method to obtain neutron spectra in the energy range from 0.3 eV to 2 keV. Preliminary resonance parameters obtained from transmission experiments below 112 eV and comparison with previous measurements are presented here. Besides these measurements, the Doppler effect has also been investigated for  $\text{NpO}_2$ . We show that the use of a harmonic crystal model gives a better fit of the resonances.

## 1. Introduction

The nucleus  $^{237}\text{Np}$  is in mass one of the most important minor actinides produced in conventional nuclear power plants: about 4 tons each year worldwide.

As for all minor actinides, the neptunium can be transmuted by neutron induced fission. After neutron capture, it becomes  $^{238}\text{Np}$  which has a fission cross section much higher than the initial  $^{237}\text{Np}$ . With a very long half-life of  $2.144 \cdot 10^6$  years,  $^{237}\text{Np}$  appears then as a good candidate for transmutation. But  $^{238}\text{Np}$ , created by neutron radiative capture, decays with a half-life of only 2.1 days to  $^{238}\text{Pu}$ . In this way, in a conventional PWR reactor more than 98 % of the introduced neptunium will become plutonium. The transmutation of neptunium is then preferable in fast reactors or hybrid reactors with high thermal fluxes and has to be considered in parallel with the transmutation of plutonium.

By determining with a better accuracy the resonance parameters of  $^{237}\text{Np}$ , this work will allow to reduce the uncertainty on the  $^{237}\text{Np}$  neutron radiative capture cross section.

The actual estimated uncertainty on this cross section is about  $\pm 10\%$  from the thermal to the fast neutron energy range. This uncertainty has neglected effects on the standard functioning of the PWR reactors but, in case of a large amount of neptunium introduced in the core of reactors, the following effects are induced:

-In a PWR reactor with 1% of neptunium-237 mixed homogeneously in a UOX core (4.5 % enriched in  $^{235}\text{U}$ ), the impact of this uncertainty is about  $\pm 0.6\%$  on the effective multiplication factor  $k_{\text{eff}}$ ,  $\pm 5\%$  on the residual quantity of  $^{237}\text{Np}$  and  $\pm 1\%$  on the production of plutonium [1]. The two last values are calculated for three years of irradiation in the reactor.

-If 3 % of  $^{237}\text{Np}$  is introduced homogeneously in a large LMFBR core (of the EFR type), the impact on the  $k_{\text{eff}}$  is  $\pm 0.3$  % and  $\pm 2.7$  % on the Na-void reactivity coefficient [2].

These impacts are not large enough to prevent preliminary calculations and parametric analysis but will be not negligible for reactors dedicated to radioactive waste transmutation. For this reason, there is a request for more precise resonance parameters of the  $^{237}\text{Np}$  [3], despite the fact that several measurements of the resonance parameters have been performed in the past [4-7].

$^{237}\text{Np}$  has several large resonances at low energies, the first one at 0.49 eV. In this region, instrumental resolution effects are negligible compared to Doppler broadening. Therefore  $^{237}\text{Np}$  is also a good candidate to study the Doppler effect in dioxyde compounds.

## 2. Experimental Setup

The total and capture cross section measurements of  $^{237}\text{Np}$  were performed using the time-of-flight technique at the Geel Linear Electron Accelerator GELINA at JRC-IRMM. The neutrons, produced via Bremsstrahlung by the electron beam hitting a rotating uranium target, were moderated by water canned in two beryllium containers (4 cm thick). The neutrons enter into the flight paths through evacuated aluminium pipes of 50 cm diameter and are confined by collimators consisting of borated wax, lead and copper. In order to absorb slow neutrons that otherwise overlap with these of the following GELINA pulse, a filter of cadmium (for the experiments at 100 Hz) or a filter of  $^{10}\text{B}$  (for the experiments at 800 Hz) was placed in the neutron beam.

### 2.1 Transmission experiments

Three  $\text{NpO}_2$  samples, with thicknesses of 0.2, 1.0 and 2.0 g/cm<sup>2</sup> of  $^{237}\text{Np}$ , have been used for the transmission measurements. For the two thinnest samples, the experiment covers the energy range from 0.3 eV to 40 eV at three different temperatures (15 K, 50 K and 290 K) in order to investigate the Doppler effect. The neutron flight distance was 26.45 m. GELINA was providing 15 ns electron bursts of 100 MeV average energy, at a repetition frequency of 100 Hz and an average beam current of 12  $\mu\text{A}$ . With a similar setup, but at 49.33 m, we measured the transmission of the thickest sample covering the energy range from 0.3 to 120 eV at 290 K. The same sample was also used for the higher energy range from 45 eV to 2.6 keV at room temperature (290 K), but with GELINA running at 800 Hz, 70  $\mu\text{A}$  and with a 1.5 ns pulse width. For all the transmission experiments, the flight path angle with respect to the normal of the moderator surface was 9°.

The  $\text{NpO}_2$  sample was mounted in an automatic sample changer at 12 m (for the experiments with the two thinnest samples) and at 23 m (for the thick sample). To determine the background, filters absorbing neutrons at specific energies (“black resonances”) were moved in and out of the beam with a sample changer controlled by the data acquisition system. In order to reduce systematic errors, these measurements sequences were recorded alternatively within a cycle lasting about one hour in total. The neutrons passing through the sample were further collimated and then detected by a NE912 lithium-glass detector. The signals from  $^6\text{Li}(\text{n},\alpha)$  reaction were processed to provide the neutron time-of-flight and then stored by the data acquisition system from the trademark FAST [8].

## 2.2 Capture experiments

For the capture measurements, two different kinds of samples have been used. The thin sample was an alloy of 90% aluminium and 10% neptunium with a thickness of  $0.02 \text{ g/cm}^2$  of  $^{237}\text{Np}$ . This sample has been used below 40 eV with GELINA running at 100 Hz. For the higher energies, the measurements were done with a sample of  $\text{NpO}_2$  with a thickness of  $1 \text{ g/cm}^2$  of  $^{237}\text{Np}$  and with GELINA running at 800 Hz.

The flight distance was 28.44 m and the angle of the flight path with respect to the normal of the moderator surface was  $0^\circ$ . The samples were placed upstream a special designed ionization chamber with a boron deposit for neutron flux measurements and between two small  $\text{C}_6\text{D}_6$  total energy detectors (NE230) placed at  $90^\circ$  in respect to the neutron beam direction. The  $\text{C}_6\text{D}_6$  detectors are dedicated to the detection of the gamma rays from the reaction  $^{237}\text{Np}(n,\gamma)^{238}\text{Np}$ . In order to obtain the number of capture events, a pulse height weighting function experimentally determined for our geometry [9], is applied to the  $\text{C}_6\text{D}_6$  detector response, making in our energy range the detector efficiency independent of the gamma decay cascade. The acquisition of the data was made in list mode with a data acquisition system developed at IRMM [10].

A major difficulty of this experiment is the high background coming from the natural radioactivity of the neptunium as shown in figure 1 giving the spectrum of the signal measured in the two  $\text{C}_6\text{D}_6$  as a function of the neutron energy.

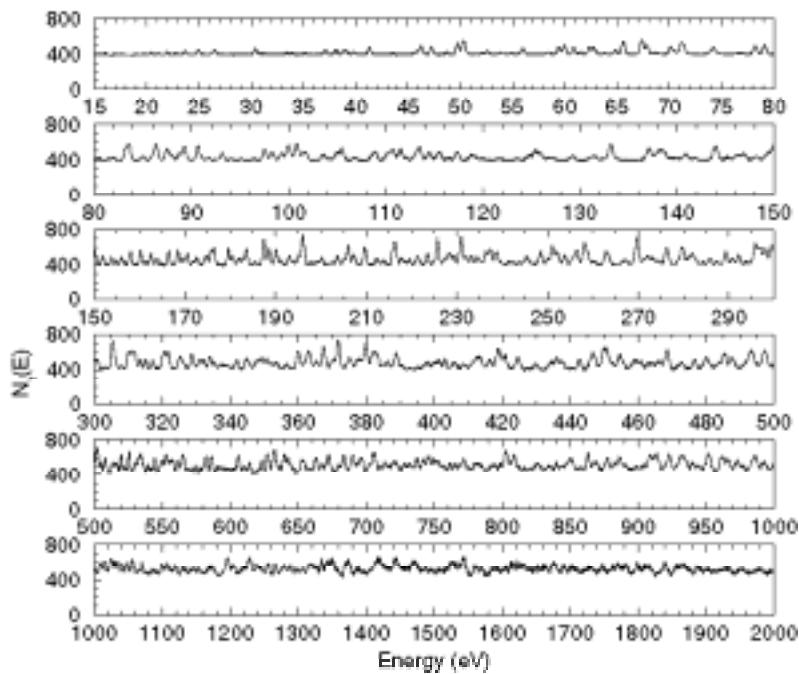


Figure 1 : Number of gamma rays in the  $\text{C}_6\text{D}_6$  detectors in function of the neutron energy  
(determined by time of flight method).

The capture data are not yet in a final form for analysis and we will just present in this paper some results from the analysis of transmission measurements.

### 3. Analysis and results of the transmission experiment

The raw time-of-flight spectra were corrected for deadtime and background. The transmission was calculated as the ratio of these corrected spectra and normalised to the ratio of the time-integrated incoming neutron fluxes of the sample “in” and “out”, measured by neutron monitors located in the target hall. The in-house developed data processing package AGS [10-11] was used to carry out the various spectrum manipulations. The experimental transmission spectrum is shown in Figure 2.

The *R*-matrix shape fitting program REFIT [12] was used in order to determine the resonance parameters: the resonance energy  $E_0$ , the neutron width  $\Gamma_n$  and the radiative width  $\Gamma_r$ . In the near future, the previous determined values of the small fission widths will be included in our analysis. It is not possible to extract from this kind of experiments the spin of the resonances. We use the spin assignments experimentally determined by Keyworth [13] for several resonances and for the other we take the values as given in the reference [14].

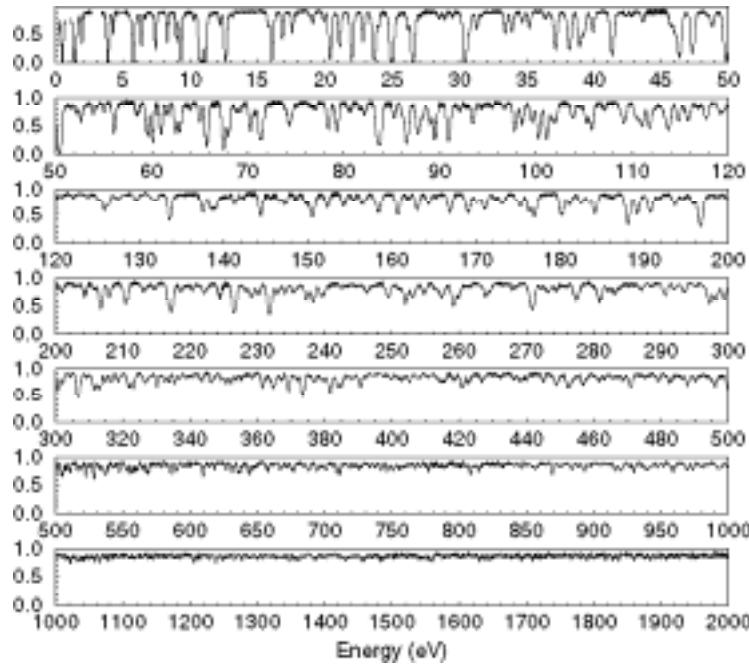


Figure 2 : Experimental transmission in function of the neutron energy between 0.3 eV and 2 keV for an  $\text{NpO}_2$  sample with  $2 \text{ g}/\text{cm}^2$  of  $^{237}\text{Np}$  placed at flight distance of 49.34 m and measured at room temperature.

We find a good agreement between our values and those of Paya [4] for the two thinnest samples, but we have a systematic discrepancy of about 3% with the thick sample. This may come from an error on the quantity of neptunium in the thick sample and the isotopic composition of this sample is currently investigated.

Therefore we give in table 1 the energies of all the resolved resonances below 112 eV, but only the values of  $\Gamma_n$  and  $\Gamma_r$ , coming from the analysis of the two thinnest samples, assuming that the fission width may be neglected and assuming an orbital momentum  $l = 0$ . These values are then still preliminary and demand further investigations. They are presented in table-1 as follows:  $E$  is the energy of the resonances and  $\Delta E$  the error on this energy including the standard deviation resulting from the fit by REFIT and an additional uncertainty of about 0.04 % of the energy due to the uncertainties on the resonance energies of  $^{238}\text{U}$  used to determinate the flight path length.  $J$  is the spin of the resonances whose values enclosed by square brackets indicate that they are assumed or preferred (see reference

[14]).  $\Gamma_n$  and  $\Gamma_\gamma$  are the preliminary values of the neutron and radiative widths. The fixed values of  $\Gamma_\gamma$  are set to 40 meV, corresponding to the mean value of the radiative width determinated by Paya [4].  $\Delta\Gamma_n$  fit and  $\Delta\Gamma_\gamma$  fit are the standard deviation on the partial widths resulting from the fit by REFIT.  $\Delta\Gamma_n$  and  $\Delta\Gamma_\gamma$  are estimations of the error including the previous standard deviation and uncertainties coming from normalization procedure and background determination.

Due to some uncertainties on the thicknesses of the samples and sample holder, a systematic error of about 1 % on the total cross section has also to be taken in account.

#### 4. Comparison with the previous experiments

The best agreement of our results are with the data of Paya [4], except for the 5 first resonances.

We confirm also the presence of a great part of the resonances only reported by Auchampaugh [7]. This experiment had better resolution (higher flight path length, low temperature) and a thicker sample than in the case of Paya but also in comparison with our experiments. In figure 3 is shown the cumulative number of resonances as a function of the energy for the measurements of Paya (used in the JEF2.2 nuclear data files for  $^{237}\text{Np}$  [15]), Auchampaugh (whose values are also given in reference [14]) and for our measurements.

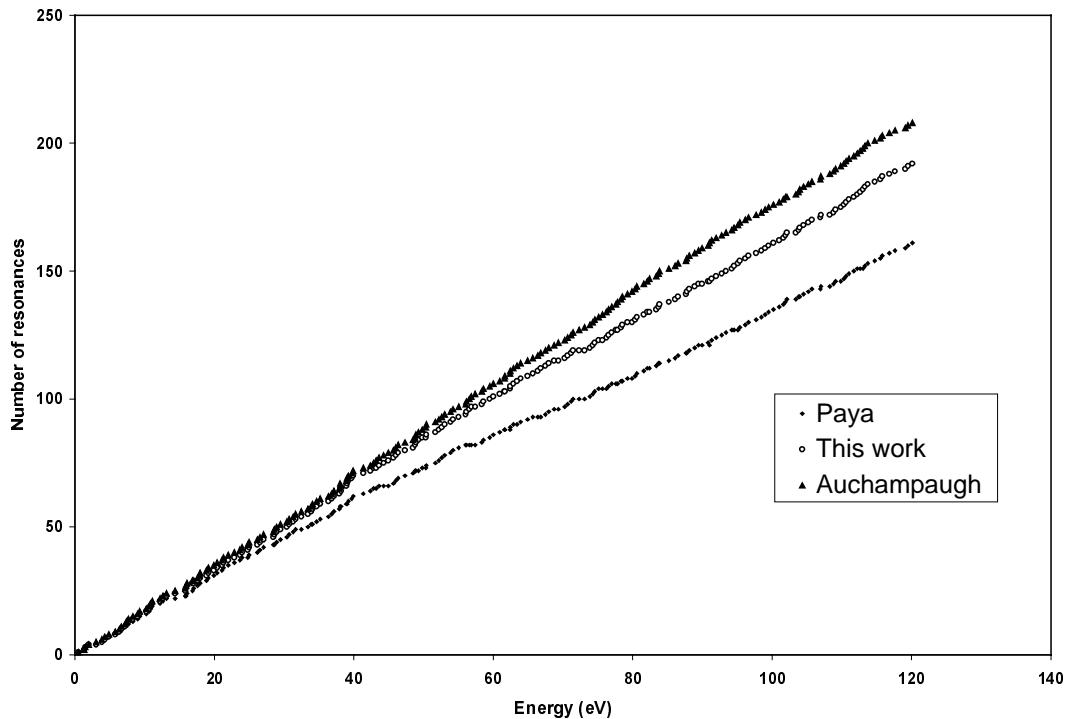


Figure 3 : Cumulative number of resonances in function of neutron energy for our experiment and comparison with the measures of Paya [4] and Auchampaugh [7].

We have found 85 resonances below 50 eV and with a linear regression in this range we obtain a value for the mean level spacing of  $0.58 \pm 0.03$  eV which is in good agreement with the value of  $0.56 \pm 0.05$  eV given by the JEF evaluation [15]. Our data are not yet analysed using a missing level estimator in order to be compared to the two values given by Auchampaugh:  $0.56 \pm 0.01$  eV or  $0.51 \pm 0.03$  eV [7].

Above 50 eV, although the agreement with Paya's values is still good (see figure 4), Auchampaugh's values of the neutron widths diverge from ours as shown in figure 5. In these figures, we represent the ratio of  $g\Gamma_n$  (with  $g$  the spin statistical weight factor) for selected resonances as a function of the neutron energy. We excluded all resonances from unresolved doublets reported by at least one author and resonances determined with low accuracy.

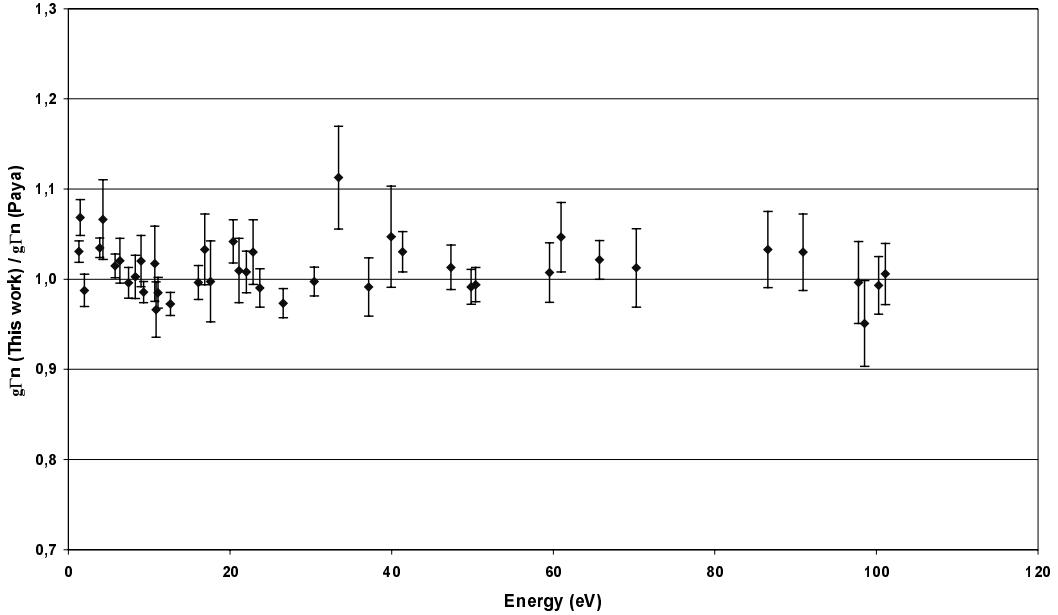


Figure 4 : Ratio between the values of neutron width determinated in our experiments and the value of Paya [4] (used in JEF2.2 [15]) for selected resonances. The error is determined from the total uncertainty on each neutron width.

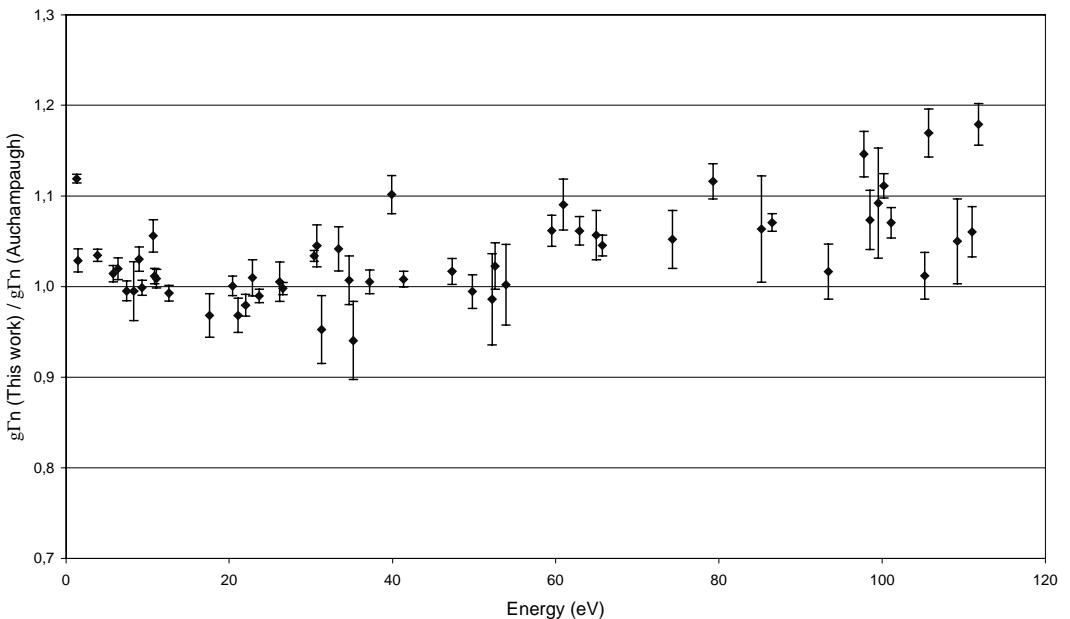


Figure 5 : Ratio between the neutron width determinated in our experiments and the value of Auchampaugh [7] (used in reference [14]) for selected resonances. The error is determined from the total uncertainty on each neutron width.

## 5. Doppler Effect Study

At low energy, the major component of the broadening of the resonances for our measurements is the Doppler effect. In the code REFIT, the Doppler broadening of the resonances is taken into account by the use of the free gas model with an effective temperature instead of the thermodynamic temperature, according to Lamb's theory [16]. Although this model reproduces well the shape of the resonances at room temperature and above 10 eV or so, this is not the case at lower energy or low temperatures, where Lamb's approximation for free gas model is no more valid.

In a previous communication [17], we mentioned that the use of the free gas model for Doppler broadening does not allow to reproduce exactly the shape of the resonances at low energy and room temperature. This effect is clearly visible in the shape of the residual of the fit which is the ratio (fit-data)/ $\sigma_{\text{data}}$ .

By using the program DOPUSH developed by D. Naberejnev [18] which describes the Doppler broadening with a more realistic model derived from the Lamb's crystal model, we showed that the shape of the cross sections generated by DOPUSH was very similar to the experimental one. The main hypothesis in this study is the use of the UO<sub>2</sub> phonon spectrum instead of the one of NpO<sub>2</sub> which is unknown. Because UO<sub>2</sub> and NpO<sub>2</sub> have similar mass and crystal structure, this assumption appears quite reasonable.

The difference between the two descriptions for Doppler broadening is mainly expected at low energy and low temperature. For this reason, we compare the Doppler broadening of DOPUSH to experimental transmission data at 4 eV (where the total width of the resonance is mainly due to Doppler broadening) and at a temperature of 50 K.

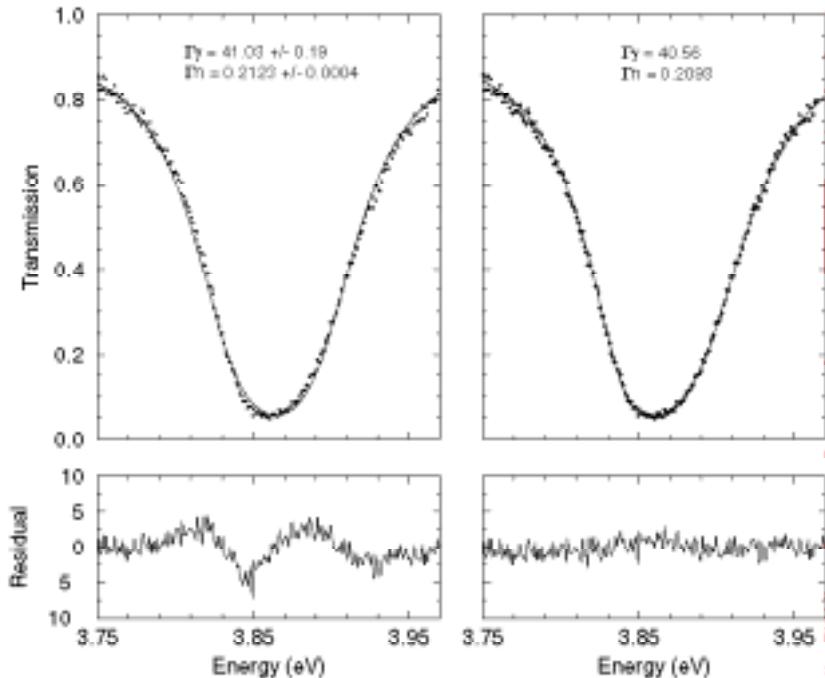


Figure 6 : Comparison between the experimental transmission at 50 K for the 3.85 eV resonance of <sup>237</sup>Np and at left, the fit by REFIT (using the gas model for Doppler broadening), and at right the simulated transmission using the harmonic crystal model of DOPUSH. The residual is the difference between the curve (fit or simulated transmission) and the data normalized by the standard deviation of the data. The uncertainties on the parameters (in meV) are only the standard deviation resulting from the fit. The values of the widths and standard deviations resulting from the fit differ from those of table-1 which were determined from data at room temperature.

A simulated transmission was created from the total cross section Doppler broadened with the harmonic crystal model of DOPUSH. By adjusting the resonance parameters, we could match very closely the experimental data as shown in figure 6 on the right. A least square fit with the free gas model using REFIT shows less agreement with the experimental points as shown in figure 6 on the left.

The inadequate description given by the free gas model for the Doppler broadening appears clearly in transmission experiments at the peak and in the wings of the resonances.

It is foreseen to implement the harmonic crystal model from DOPUSH for Doppler broadening into REFIT.

## 6. Conclusion

The preliminary results of the transmission analysis are in good agreement with the values of Paya [4] used in all the nuclear data files for resonance parameters above 1 eV. For the first resonance, our values are close to those determined in the JEF evaluation [15]. We resolved also several doublets. Our experimental data does not allow us to confirm the existence of all the resonances reported by Auchampaugh [7]. The analysis is still on going and final results for the resonance parameters up to 600 eV and mean values from 600 eV to 2 keV are foreseen later on.

The description of the Doppler broadening by Lamb's harmonic crystal model used in DOPUSH provides a good description of the shape of the resonances even at low energies or low temperatures (50 K). In these conditions, the R-matrix least square analysis program which uses the free gas model for Doppler broadening gives an inadequate shape of the resonances. Values for the resonance parameters, especially the radiative widths, are then biased. These values will be corrected in the near future by implementing the DOPUSH description for Doppler broadening in REFIT.

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Table 1 : List of preliminary resonance parameters (Energy in eV and widths in meV) with standard deviation resulting from the fit and estimation of total error.

E	$\Delta E$	J	$\Gamma n$	$\Delta \Gamma n$ fit	$\Delta \Gamma n$	$\Gamma \gamma$	$\Delta \Gamma \gamma$ fit	$\Delta \Gamma \gamma$	E	$\Delta E$	J	$\Gamma n$	$\Delta \Gamma n$ fit	$\Delta \Gamma n$	$\Gamma \gamma$	$\Delta \Gamma \gamma$ fit	$\Delta \Gamma \gamma$
-1.000		3	0.922	0.106	0.129	40,00			27,11	0,02	(2)	0,042	0,007	0,009	40,00		
-0.613		2	0.239	0.065	0.072	40,00			28,46	0,01	2	0,091	0,007	0,007	40,00		
0,4918	0,0002	2	0,04790	0,00004	0,00099	40,52	0,07	0,39	28,61	0,02	(3)	0,036	0,004	0,004	40,00		
1,321	0,001	3	0,0327	0,0001	0,0002	38,78	0,17	0,27	28,93	0,01	2	0,132	0,007	0,008	40,00		
1,478	0,001	2	0,1859	0,0002	0,0023	42,05	0,10	0,34	29,48	0,01	2	0,083	0,008	0,008	40,00		
1,968	0,001	3	0,0144	0,0001	0,0001	37,41	0,59	0,60	30,42	0,01	3	3,215	0,013	0,026	44,06	1,21	1,48
3,864	0,002	3	0,2164	0,0004	0,0019	39,81	0,20	0,48	30,75	0,01	2	0,381	0,007	0,008	40,00		
4,264	0,002	2	0,0333	0,0005	0,0005	40,47	1,88	1,88	31,30	0,01	3	0,239	0,012	0,013	37,19	13,92	14,52
4,860		2							31,66	0,02	3	0,041	0,007	0,007	40,00		
5,777	0,002	3	0,541	0,001	0,007	42,74	0,22	0,86	32,49		2						
6,376	0,003	3	0,081	0,001	0,001	40,54	1,17	1,83	33,42	0,01	3	0,418	0,012	0,014	42,69	8,65	9,36
6,679	0,003	2	0,013	0,001	0,001	40,00			33,90	0,01	2	0,507	0,010	0,011	40,00		
7,192	0,004	2	0,010	0,001	0,001	40,00			34,08	0,03	3	0,037	0,007	0,008	40,00		
7,421	0,003	3	0,125	0,001	0,002	38,28	1,09	1,58	34,69	0,01	3	0,172	0,006	0,006	40,00		
7,679	0,012	2	0,002	0,001	0,001	40,00			35,20	0,01	2	0,396	0,017	0,018	21,99	11,35	11,85
8,304	0,003	3	0,092	0,001	0,002	37,90	1,65	2,69	36,39	0,02	3	0,131	0,008	0,009	40,00		
8,976	0,004	3	0,106	0,001	0,002	40,01	1,82	2,43	36,82	0,02	(2)	0,118	0,009	0,010	40,00		
9,297	0,004	2	0,617	0,002	0,004	42,16	0,53	0,78	37,15	0,01	3	1,156	0,017	0,020	37,07	4,69	5,02
10,227	0,005	2	0,030	0,001	0,002	40,00			37,84	0,03	(2)	0,053	0,011	0,023	40,00		
10,680	0,004	3	0,441	0,005	0,007	37,54	1,25	1,53	38,04	0,02	2	0,172	0,018	0,018	40,00		
10,844	0,004	3	0,729	0,007	0,009	41,23	1,32	1,48	38,19	0,02	3	1,235	0,040	0,041	40,00		
11,096	0,004	2	1,046	0,004	0,010	42,70	0,73	0,95	38,92	0,02	3	0,917	0,041	0,042	40,00		
12,200		3							39,02	0,02	2	0,332	0,053	0,054	40,00		
12,616	0,005	2	0,928	0,004	0,008	40,51	0,73	0,96	39,24	0,02	3	0,541	0,009	0,009	40,00		
13,128	0,007	3	0,018	0,001	0,002	40,00			39,78	0,09	2	0,024	0,020	0,020	40,00		
14,403	0,168	(2)	0,003	0,003	0,004	40,00			39,92	0,02	3	0,499	0,012	0,013	40,00		
15,789	0,007	3	0,071	0,002	0,002	40,00			41,36	0,02	3	1,996	0,018	0,024	37,13	2,93	3,44
15,912	0,008	(3)	0,025	0,002	0,002	40,00			42,37	0,02	3	0,090	0,010	0,010	40,00		
16,086	0,006	2	1,105	0,007	0,013	46,10	1,34	1,82	42,80	0,02	(3)	0,083	0,012	0,012	40,00		
16,857	0,007	2	0,301	0,005	0,007	38,78	3,31	4,35	43,63		2						
17,592	0,007	3	0,157	0,004	0,005	31,49	5,23	6,30	44,29		2						
17,907	0,013	2	0,019	0,004	0,004	40,00			44,94		2						
18,88		2							45,70		2						
19,12		3							46,01		3						
19,93	0,01	3	0,067	0,003	0,004	40,00			46,34		3						
20,40	0,01	2	1,413	0,007	0,015	47,48	1,24	1,81	47,33	0,02	2	2,918	0,037	0,041	35,92	4,48	4,50
21,10	0,01	(3)	0,447	0,003	0,004	34,17	3,62	4,79	48,46	0,03	(2)	0,131	0,017	0,018	40,00		
21,28	0,01	2	0,035	0,006	0,006	40,00			48,78	0,02	3	0,435	0,015	0,015	40,00		
22,01	0,01	2	1,524	0,008	0,017	42,72	1,26	1,97	48,98	0,03	2	0,134	0,023	0,027	40,00		
22,86	0,01	3	0,395	0,006	0,011	44,01	3,60	5,26	49,33	0,07	(2)	0,029	0,016	0,023	40,00		
23,67	0,01	3	1,435	0,010	0,015	40,62	1,55	1,84	49,82	0,02	3	4,326	0,031	0,045	35,50	2,60	2,60
23,99	0,01	2	0,197	0,014	0,016	52,53	15,88	17,65	50,40	0,02	3	7,608	0,033	0,066	48,27	1,82	2,12
24,78	0,01	3	0,045	0,003	0,004	40,00			51,69	0,03	3	0,095	0,013	0,015	40,00		
24,98	0,01	3	3,589	0,011	0,048	40,00			52,21	0,02	2	0,420	0,019	0,021	40,00		
26,19	0,01	3	0,206	0,005	0,006	40,00			52,64	0,02	2	0,895	0,019	0,022	40,00		
26,56	0,01	3	2,369	0,009	0,022	46,69	1,08	1,62	53,06	0,03	3	0,063	0,013	0,015	40,00		
53,87	0,02	2	0,509	0,020	0,022	40,00											
54,24	0,03	(2)	0,206	0,022	0,025	40,00											
55,04	0,02	3	0,305	0,014	0,016	40,00											
56,01	0,02	2	1,268	0,096	0,099	40,00											
56,15	0,02	3	0,706	0,066	0,073	40,00											
56,49	0,04	(2)	0,082	0,022	0,023	40,00											
56,87		3															
58,40	0,02	(3)	0,389	0,020	0,022	40,00											
58,64	0,03	3	0,239	0,019	0,022	40,00											
59,51	0,02	2	2,430	0,032	0,034	40,00											
60,02		3															
60,96	0,02	3	1,642	0,042	0,049	46,73	10,51	11,34									
61,64	0,03	(3)	0,462	0,018	0,029	40,00											
62,34	0,06	2	0,238	0,125	0,129	42,25											
62,49	0,03	3	1,581	0,092	0,093	43,23											
62,91	0,03	3	1,501	0,022	0,030	40,00											
63,40	0,06	(2)	0,074	0,025	0,038	40,00											
63,96	0,03	(3)	0,256	0,019	0,029	40,00											
64,97	0,03	3	0,901	0,021	0,031	40,00											
65,71	0,03	3	3,984	0,045	0,061	48,26	5,83	6,88									
66,40		2															
66,80		(3)															
67,46		3															
67,94		2															
68,79	0,03	3	0,353	0,025	0,025	40,00											
70,26	0,03	3	1,857	0,032	0,034	40,00											
70,66		2															
71,20	0,03	3	1,998	0,038	0,059	40,00											
71,46	0,03	2	3,203	0,057	0,064	40,00											
73,87	0,03	3	0,292	0,026	0,027	40,00											
74,29	0,03	2	1,757	0,052	0,053	40,00											
74,58	0,03	3	0,411	0,032	0,033	40,00											
75,20	0,05	(2)	0,135	0,02													

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