NEUTRON RESONANCES IN PRASEODYMIUM AND TERBIUM

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Submitted to JETP editor January 28, 1964

J. Exptl. Theoret. Phys. (U.S.S.R.) 47, 43-51 (July, 1964)

The resonance parameters of Tb and Pr, including the spin values for a number of levels, were determined by measuring the transmission, radiative capture, and scattering of neutrons. For Tb, 22 levels up to 100 eV were analyzed; for Pr, 14 levels up to 1000 eV. Four levels were observed for the first time. A mean radiation width of 86 MeV was found for both Tb and Pr levels.

HIS article reports new measurements that were obtained using a neutron time-of-flight spectrometer and the pulsed fast reactor of the Joint Institute for Nuclear Research. The first measurements, on rhodium and bromine, were published in ^[1,2]. The rare earth elements terbium and praseodymium were selected for the continuation of the investigation. The data for these elements in the atlas of neutron cross sections ^[3] do not go beyond 1955 and do not include the spins and values of Γ_{γ} for individual resonances. Only one recent publication ^[4] gives results for praseodymium obtained by the transmission method. However, with regard to the radiation widths most of the data in ^[4] cannot be regarded as even a rough estimate.

In the work on praseodymium and terbium we expected to obtain the radiation widths and spins of several levels and to refine the distributions of the reduced neutron widths of these elements, especially for praseodymium, for which, according to ^[4], the number ν of degrees of freedom in the distribution of reduced neutron widths was six instead of $\nu = 1$ as in the generally assumed Porter-Thomas distribution.

1. TRANSMISSION MEASUREMENT

The apparatus described in ^[5] was used to measure transmission. The transmission of terbium was measured at energies from 3 to 80 eV using samples of terbium oxide having 1.22 $\times 10^{20}$, 6.94 $\times 10^{20}$, 2.76 $\times 10^{21}$, and 1.498 $\times 10^{22}$ Tb nuclei per cm². Praseodymium was measured from 8 to 1700 eV; the thicknesses of the praseodymium oxide samples were 1.08×10^{21} , 3.25×10^{21} , 9.42×10^{21} , 2.355×10^{22} , and 7.3×10^{22} Pr nuclei per cm². The experimental results were treated by the method described in ^[1]. This method is based on determining experimentally the area A of the dip of the transmission curve; this transmission dip area is a function of the resonance parameters and sample thickness n (nuclei/cm²):

$$A = A \left(g \Gamma_n, \Gamma, n \right). \tag{1}$$

2. RADIATIVE CAPTURE MEASUREMENT

The radiative capture of neutrons was studied using the detector described in ^[6]. Two alternating series of measurements were obtained. In the first series only one sample D was placed in the neutron beam directly in the detector channel. It was shown in ^[2] that in this case we obtain

$$\Sigma N(D) / \Pi(E_0) \varepsilon_{\gamma} = A_D \Gamma_{\gamma} / \Gamma = C, \qquad (2)$$

where $\Sigma N(D)$ is the sum of detector counts over the entire resonance, and the product of the total flux $\Pi(E_0)$ and registration efficiency of radiative capture ϵ_{γ} is determined experimentally by one of the methods described in ^[7].

In the second series of measurements a second sample T (containing n_T nuclei per cm²) was placed in the beam path at a considerable distance from the detector. From the ratio of the sum of counts for the entire resonance in the presence and absence of the transmitting sample T we obtain

$$\Sigma N(D,T) / \Sigma N(D) \exp(n_T \sigma_p) = (A_{D+T} - A_T) / A_D.$$
 (3)

This method of self-indication was described in detail in [8,2].

It should be noted that in the case of resonances with $\Gamma_n \gg \Gamma_{\gamma}$ dependence on the parameters Γ_n and Γ practically disappears from the right-hand side of (2). This is easily shown by substituting in (2) the analytic expression for A_D (see ^[9], for example):

$$A_{D} = \frac{\pi}{2} n \sigma_{0} \Gamma e^{-n \sigma_{0}/2} \left[I_{0} \left(n \sigma_{0}/2 \right) + I_{1} \left(n \sigma_{0}/2 \right) \right].$$

Since in this case $\Gamma_n/\Gamma \sim 1$ and $\sigma_0 \sim g\Gamma_n/\Gamma$, only the dependence on Γ_γ and g remains in the right-hand side of (2). This means that for a given value of g the radiation widths of the resonances with $\Gamma_n \gg \Gamma_\gamma$ can be determined from resonance capture measurements independently of the exact values of Γ_n and Γ . This situation is not changed by taking Doppler broadening into account. The radiation widths of most Pr resonances were obtained in this manner.

Equations (2) and (3) were introduced by assuming single interactions of neutrons with nuclei of the samples. However, with samples of intermediate thicknesses, especially in the case $\Gamma_n \gg \Gamma_{\gamma}$, C_{exp} included detector counts associated with neutron capture in a second or later interaction, and counts associated with the registration of scattered neutrons. When using (2) we therefore introduced a correction defined by

$$C = C_{\exp} \left(1 - Q_2 / Q_1 \right) \left(1 + \Gamma_n \varepsilon_n / \Gamma_{\gamma} \varepsilon_{\gamma} \right)^{-1}.$$
 (4)

The correction factor $1 - Q_2/Q_1$ required by secondary interactions contains the functions Q_1 and Q_2 of the resonance and thickness parameters. Q_1 and Q_2 are defined as the probabilities of first and second interactions of a neutron in the sample and were calculated in ^[10]. The correction $(1 + \Gamma_n \epsilon_n / \Gamma_\gamma \epsilon_\gamma)^{-1}$ associated with the registration of scattered neutrons requires no explanation.

The measurements of the radiative capture of neutrons by praseodymium and terbium nuclei were obtained with a 750-meter flight path and $0.05-\mu$ sec/m resolution. The samples were oxides of the investigated elements filling thin-walled aluminum cassettes 190 mm in diameter. For Pr we used four D samples having thicknesses from 1×10^{21} to 5.5×10^{21} nuclei/cm², and four T samples having from 8×10^{20} to 2.1×10^{22} nuclei/cm². For Tb we used two D samples with 7.9×10^{20} and 15.3×10^{20} nuclei/cm², and one T sample with 7.4×10^{20} nuclei/cm².

3. NEUTRON SCATTERING MEASUREMENT

In our investigation of neutron scattering the directly measurable quantity was the difference between the experimental sum ΣN of counts in the resonance region and the sum ΣN_p of counts in the same channels resulting only from potential scattering in the absence of resonance interactions. This difference is associated with the resonance parameters as follows:

$$\frac{\Sigma N - \Sigma N_p}{\Pi (E_0) \varepsilon_n} = \frac{\Gamma_n}{\Gamma} A (1 - \Omega) f.$$
(5)

Equation (5) without the factor f has been given in ^[2]. Here $1 - \Omega$ is a correction factor, thoroughly discussed in ^[2], which is required by the fact that the resonance interaction of neutrons reduces the number of neutrons undergoing potential scattering. The product of the total flux and the neutron registration efficiency $\Pi(E_0)\epsilon_n$ of the detector is obtained from calibration measurements with lead, as is shown in ^[2]. The correction factor f takes into account neutron capture after scattering. For a detector with the constant efficiency ϵ_n , which is independent of neutron energy and emission angles, this factor can be represented by

$$f = \frac{1 - (\Gamma/\Gamma_n) (Q_2/Q_1)}{1 - Q_2/Q_1}, \qquad (6)$$

where Q_1 and Q_2 have the same meaning as in (4). The correction factor can be utilized whenever the procedure of extrapolating to zero sample thickness, as described in ^[2], cannot be adopted.

It should be noted that in the cases of resonances with $\Gamma_n \gg \Gamma_\gamma$ the value of A customarily obtained from transmission can be determined by scattering measurements. The necessary condition for this purpose is the independent determination of Γ_γ , thus enabling us to determine the ratio Γ_n/Γ with 1–2% accuracy over a broad range of Γ_n . Then (5) determines A, and the corrections in (5) in the case $\Gamma_n \gg \Gamma_\gamma$ become small. This method of determining A from scattering measurements was used for thin Pr samples, for which it was difficult to obtain highly accurate transmission measurements.

Neutron scattering was studied using the detector described in ^[11]. In the measurements with terbium the resolution was 0.08 μ sec/m with a 500-meter flight path. Three terbium oxide samples having the thicknesses 1.14×10^{20} , 1.9×10^{20} , and 7.4×10^{20} nuclei/cm² were used. In the measurements with praseodymium the resolution was 0.05 μ sec/m with a 1000-meter flight path. The praseodymium sample thicknesses were 2.02×10^{20} , 1.48×10^{21} , and 1.66×10^{21} nuclei/cm².

4. RESULTS AND DISCUSSION

Equations (1)-(3) and (5) were used to plot the dependence of $g\Gamma_n$ on Γ for each measured sample. For this purpose the tables computed in ^[12] were used. Equations (2) and (5) each gave two dependences according to the values assumed for the statistical factor g. One of the two families of curves obtained by this procedure should be

Table I. Parameters of Tb resonances

| E_0 , eV | $g\Gamma_n$, MeV | Γ_{γ} , MeV | J | $2g\Gamma_n^0$ |
|--------------------------------------|--------------------------------------|-------------------------|----------------|----------------|
| 3 340 ± 0 005 | 0.21 ± 0.02 | 80 ± 10 | 9* | 0.23 |
| 3.340 ± 0.003 | 0.21 ± 0.02 0.032 ± 0.003 | 00 I 10 | 1* | 0.029 |
| 4.50 ± 0.01 41.05 ± 0.02 | 5.0 ± 0.2 | 87 + 6 | 2 | 3 02 |
| $11,03 \pm 0.02$ $14,38 \pm 0.03$ | 0.084 ± 0.008 | 01 ± 0 | 4. | 0,044 |
| 14.00 ± 0.00 21.20 ± 0.05 | 0.034 ± 0.000 | | | 0.26 |
| 24.6 ± 0.07 | 27 ± 0.00 | 76 ± 11 | 2 | 0.12 |
| 27.6 ± 0.08 | 0.47 ± 0.05 | 70 <u>r</u> 11 | - | 0.18 |
| 33.9 ± 0.11 | 1.4 ± 0.2 | 81 + 15 | 1 | 0 48 |
| 40.7 ± 0.15 | 0.29 ± 0.05 | | - | 0.091 |
| 43.7 ± 0.17 | 2.5 ± 0.2 | 76 + 16 | 2 | 0.765 |
| 46.1 ± 0.18 | 6.7 ± 0.5 | 89 + 11 | $\overline{2}$ | 1.97 |
| 50.3 ± 0.20 | 1.3 + 0.3 | | _ | 0.37 |
| 51.6 + 0.20 | 0.54 + 0.07 | | | 0.15 |
| 54.2 ± 0.23 | 0.30 + 0.05 | | 1 | 0,081 |
| 57.3 ± 0.25 | 0.66 ± 0.07 | | | 0.176 |
| 58.7 ± 0.25 | 0.85 ± 0.15 | | | 0,22 |
| 65.5 ± 0.30 | 5.0 ± 0.5 | 92 ± 20 | | 1,25 |
| 66.8 ± 0.30 | 1.2 ± 0.1 | _ | | 0,29 |
| 74.0 ± 0.35 | 8.5 ± 0.6 | 87 ± 16 | 2 | 1.98 |
| 76.8 ± 0.4 | | | | |
| 78.0 ± 0.4 | | | | |
| 88.8 ± 0.5 | 1.8 ± 0.2 | | | 0.38 |
| 90.6 ± 0.5 | 3.8 ± 0.3 | | | 0.8 |
| 97.5 ± 0.5 | 14 ± 1 | 103 ± 14 | 1 | 2.7 |

*Spins given in^[13].

contradictory in virtue of an incorrect selection of g. In our present work this was the family of curves, obtained from (2) and (5), which passed the effective intersection center of the other curves at a distance exceeding two standard errors, while at the same time the analogous curves associated with the other value of g passed the effective center within the limits of one standard error.

<u>Terbium</u>. The measurements obtained with terbium are given in Table I. The spectrometer resolution permitted an analysis of 22 levels up to 100 eV, without taking into account the poorly resolved 76.8- and 78-eV resonances.

For all resonances the data on the radiative capture of neutrons by terbium nuclei were calibrated on the basis of the count at the maximum of the strong and well-resolved 11.1-eV resonance by the method described in ^[7].

The complete program of treatment was applied to the resonances for which the level spins J are given in the table. Scattering data were extrapolated to zero sample thickness using three points (three samples), for resonances at 11.1, 24.6, 46.1, and 97.5 eV. If the strongest resonance at 11.1 eV is excluded, the extrapolation leads to the same results as the correction procedure employing (6). Consequently, for the other three weaker levels the scattering data were obtained from (5) and corrected according to (6). The highest correction was 10%.

For the remaining resonances, having smaller neutron widths, scattering measurements obtained by the self-indication method were not sufficiently accurate. For these resonances we therefore obtained only values of $g\Gamma_n$ based on the assumption $\Gamma = \Gamma_{\gamma} = 86$ MeV.

We determined the spins of seven levels. A polarization technique was recently used in $\begin{bmatrix} 13 \end{bmatrix}$ to determine the spins of resonances at 3.44, 4.98, and 11.1 eV. The spin values agreed for the 11.1eV resonance. We did not determine the spins for the 3.34- and 4.98-eV resonances because of their very small neutron widths.

It follows from Table I that the number of levels with a given spin value agrees with the known level density $\rho(J) \sim 2J + 1$. Within the limits of experimental error the radiation width is not observed to depend on the spin of an excited nuclear state. Also, there are no appreciable fluctuations of the radiation width from resonance to resonance. There is a noteworthy large difference between our data and those in ^[14] for $g\Gamma_n$ at energies above 50 eV; our values are considerably smaller, but a number of levels observed by us are absent from ^[14]. It is extremely likely that because of insufficient resolution two or more close-lying levels were previously regarded as a single level.

The data in Table I enabled us to obtain the values of the mean reduced neutron width $2g\Gamma_n^0 = 0.75 \pm 0.23$ MeV, the observed mean level separation (without spin splitting) $D = 4.2 \pm 0.6$ eV, and the strength function $S_0 = (0.90 \pm 0.30) \times 10^{-4}$. The indicated errors are due mainly to the statistics of the number of observed levels and were calculated assuming a Wigner distribution for the intervals between levels, and a Porter-Thomas distri-



FIG. 1. Distribution of reduced neutron widths. a – terbium, b – praseodymium. Smooth curve – Porter-Thomas distribution for $\nu = 1$.

bution of reduced neutron widths. Figure 1a shows that the latter distribution satisfactorily describes the experimental data.

Praseodymium. For praseodymium we determined the parameters of 14 resonances up to 1000 eV; the results are collected in Table II. The weak levels at 50.5, 112, 411, and 470 eV were observed for the first time. The certified purity of the samples was at least 99.5%. These resonances should be strong if they belong to impurity nuclei. A comparison was made with all strong resonances in [³], and additional measurements were obtained with neodymium, which was the most probable impurity. The noted resonances were found to be new ones belonging to praseodymium.

The values of $g\Gamma_n$ for these levels were ob-

tained by using (2), where $\Pi(\mathbf{E}_0)\epsilon_{\gamma}$ is known and Γ_{γ}/Γ is of the order of unity for weak resonances. The parameters of the 85.1- and 384-eV resonances with small neutron widths were obtained by the conventional combined analysis of the plotted curves of $g\Gamma_n$ and Γ based on (1)-(3).

The remaining resonances of praseodymium have greater neutron widths; we were thus enabled to determine their radiation widths from the measurements of radiative neutron capture. As a result, however, unlike the measurements with terbium, where all corrections were small, in the measurements with praseodymium the correction factor in (4) associated with the registration of scattered neutrons now differed from unity up to 15%, and the correction factor $1 - Q_2/Q_1$ differed from unity by 20-30%, as a rule. It should be noted that after introduction of the appropriate corrections the data regarding radiative capture for samples of different thicknesses were in agreement, giving close values of Γ_{γ} .

Since there are no praseodymium resonances with reliably measured parameters, the values of $\Pi(E_0)$ and ϵ_{γ} in (2) for praseodymium were determined separately by the method given in ^[7]. The errors in the values of the total flux and efficiency were estimated to be each not more than 10%. Since $\Sigma N(D)$ is accurate to within at least 5%, we obtain for C a total rms error of 15%. However, taking into account also the possible inaccuracy resulting from a correction for capture after scattering, we estimate the final error of Γ_{γ} at 20%.

In the case of strong resonances the curves relating $g\Gamma_n$ and Γ obtained from (2) are reduced to two lines:

$$g_i(\Gamma - \Gamma_n) = g_i \Gamma_{\gamma}, \quad i = 1, 2 \tag{7}$$

which intersect the curves obtained from (1) for transmission. The spins were obtained in the cases when the transmission and scattering curves deter-

| | E_{o} , eV | ${}_{g\Gamma_n}$, MeV | Γ _Υ , MeV | J | $2g\Gamma_n^0$, MeV |
|---|-----------------------|------------------------|----------------------|-----|----------------------|
| - | 50.5 ± 0.2 | 0.030 ± 0.005 | | | 0.0085 |
| | 85.1 ± 0.2 | 3.2 ± 0.4 | 80 + 20 | | 0,70 |
| | 112.0 ± 0.3 | 0.23 ± 0.03 | | | 0,044 |
| | 216.7 ± 0.8 | 570 ± 40 | 71 + 15 | 3 | 77.5 |
| | 234 + 1 | 460 ± 25 | 93 ± 15 | (3) | 60.2 |
| | 359 ± 2 | 790 ± 50 | 60 ± 15 | 3 | 82.9 |
| | 384 ± 3 | 27 ± 4 | 125 ± 30 | | 2.75 |
| | 411 ± 5 | 1.2 ± 0.2 | | | 0.118 |
| | 470 ± 5 | 1.7 ± 0.3 | | | 0.157 |
| | 515 <u>+</u> 4 | 380 ± 25 | 75 ± 15 | 3 | 33.8 |
| | 631 ± 5 | 1200 ± 50 | 107 ± 20 | | 95.7 |
| | 718 ± 6 | 1200 ± 60 | 68 ± 15 | _ | 89.5 |
| | 839 ± 7 | 2200 ± 100 | 83 ± 15 | 3 | 151.6 |
| | 942 + 8 | 1470 ± 70 | 107 + 20 | 1 2 | 96.0 |

Table II. Parameters of Pr¹⁴¹ resonances



FIG. 2. The curves B_1 , B_2 , and B_3 were obtained from measurements of scattering on samples having the respective thicknesses 2.02×10^{20} , 1.48×10^{21} , and 1.66×10^{21} nuclei/cm². The curves T_1 , T_2 , and T_3 were obtained from transmission measurements for samples having the thicknesses 10.8×10^{21} , 2.35×10^{22} , and 9.4×10^{21} nuclei/cm². The curves C were obtained from radiative capture measurements for sample thicknesses $(1 - 5.5) \times 10^{21}$ nuclei/cm². The assumed values of g are shown in parentheses: (7/12) and (5/12).

mined independently the set of parameters $(g\Gamma_n, \Gamma)$ that fitted one of the curves (7) within its standard error (~10% for Γ), and was two or more standard errors distant from the curve with the other spin. The determination of the parameters of strong resonances is illustrated in Fig. 2.

We determined the spins for five levels. The spin of the 234-eV resonance, which appears in parentheses in the table, follows from the inter-ference between the 216.7- and 234-eV levels which was observed in ^[15]. Thus only one level (942 eV) of the six has spin J = 2.

With regard to the radiation widths it should be noted that there are no fluctuations from level to level, within the limits of experimental error.

An analysis of the data in Table II leads to the following values of the mean characteristics of praseodymium nuclei:

$$\begin{split} \overline{2g\Gamma_n^0} &= (49 \pm 18) \,\,\mathrm{MeV}\,, \qquad D = (64 \pm 13) \,\,\mathrm{eV}\,, \\ \overline{\Gamma}_{\gamma} &= 86 \,\,\mathrm{MeV}\,, \quad S_0 = (3.6 \pm 1.4) \cdot 10^{-4}. \end{split}$$

It is interesting to compare the value of $\overline{\Gamma}_{\gamma}/D_0$ obtained in ^[16] by measuring the mean cross section for radiative capture below 50 keV, with our present results. Remembering that $D_0 = D$ $\times 2(2I + 1)$, our data give $\overline{\Gamma}_{\gamma}/D_0 = (1.1 \pm 0.3)$ $\times 10^{-4}$, whereas ^[16] gives $(0.60 \pm 0.07) \times 10^{-4}$. This discrepancy is almost within the measurement error, which is large in our case and is associated to a considerable degree with the small statistics of the levels. However, there is another possible explanation. Table II shows that the reduced widths of the levels at 50, 112, 411, and 470 eV are appreciably smaller than those of the other levels. If these levels are attributed to p neutrons, D₀ is augmented and the values of $\overline{\Gamma_{\gamma}}/D_0$ are in good agreement. The hypothesis that the noted resonances involve p waves does not conflict with the value obtained therefrom for the strength function S₁ = $(2.5 \pm 2.0) \times 10^{-4}$. This is consistent with the value S₁ = $(1.1^{+1.1}_{-0.6}) \times 10^{-4}$ obtained in ^{[16}].

The distribution of neutron widths for all 14 levels is given in Fig. 1b along with the Porter-Thomas distribution for $\nu = 1$. The agreement is not good; however, the Kolmogorov probability test gives a value of about 50% for the probability of such random deviation, i.e., the experimental and theoretical distributions do not conflict. If the four weak levels are assigned to p waves there is even poorer agreement between the distributions, but in this case the Kolmogorov test gives about 25% probability for the discrepancy.

The neutron width distribution would be further refined by measurements with better resolution in a considerably broader energy range.

In conclusion we take pleasure in thanking F. L. Shapiro for his interest and for valuable discussions. We are also indebted to I. I. Shelontsev and N. Yu. Shirikova for electronic computer calculations.

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