RESONANCE PARAMETERS FOR URANIUM 233

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Summary

Single-level and multilevel resonance parameters are given for 233 U below 11 eV. The total and fission cross sections were fitted simultaneously and good agreement was obtained. Spin assignments determined by the least squares method support the prediction of the channel theory of fission that 233 U has two level populations; one with predominantly large fission widths and the other with much narrower widths.

I. INTRODUCTION

The object of the present work was firstly to obtain a set of best fitting resonance parameters for the total and fission cross sections for neutrons on 233 U, and secondly to attempt to determine resonance spin assignments by the least squares method. The most recent single-level fits of Nifenecker (1964) and Nifenecker and Perrin (1965) were performed on the fission cross section alone and give poor agreement with the total cross section, while the recommended parameters of Stehn *et al.* (1965) give poor agreement with both total and fission cross sections.

Other multilevel fits (Moore and Reich 1960; Pattenden and Harvey 1960; Vogt 1960) assign resonance spins subjectively—narrow resonances are placed in one spin grouping while broad resonances are placed in the other. This procedure results in far too many resonances in the narrow group, contrary to the expectation that level spacings for the two sequences will be approximately equal. In particular, the two-channel analysis of Moore and Reich (1960) places all resonances in the one group.

The possibility exists that many broad levels are unresolved and contribute a residual background cross section. If so, the three multilevel fits referred to are presumably in error. A calculation by Musgrove (1967) indicates that approximately one-quarter of the resonance levels in 233 U are undetected, either because they are too weak or because they overlap other levels. Accordingly, the fullness below the $3 \cdot 6 \text{ eV}$ resonance is ascribed to an overlapping level at $3 \cdot 4 \text{ eV}$ while a broad resonance is included at $6 \cdot 6 \text{ eV}$ to improve the fit in this region. The effect of adding further levels was investigated, but no significant improvement in the fit was achieved with levels having realistic parameters.

Resonance parameters below 11 eV were obtained by the least squares method, both the total and fission cross sections being fitted simultaneously to the Breit-Wigner single-level approximation and to an approximate form of the multilevel formula (Feshbach, Porter, and Weisskopf 1954).

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The single-level parameters quoted here are an improvement over those given in an earlier report (Musgrove, unpublished data 1966), where the radiation widths were assumed constant. The overall fit has been improved and, in particular, better agreement with the thermal cross sections has been achieved.

II. SINGLE-LEVEL FIT

The Breit–Wigner single-level formulae for the various cross sections are given by

$$\sigma_{\rm s} = \sigma_{\rm p} + \sigma_0 \frac{\Gamma_{\rm n}}{\Gamma} \frac{1 + (2\pi/\lambda_{\rm r}) 2R(\Gamma/\Gamma_{\rm n})X}{1 + X^2}, \qquad (1)$$

$$\sigma_{\rm c} = \sigma_0 \frac{\Gamma_{\gamma}}{\Gamma} \left(\frac{E_{\rm r}}{E}\right)^{\frac{1}{2}} \frac{1}{1+X^2},\tag{2}$$

$$\sigma_{\rm f} = \sigma_0 \frac{\Gamma_{\rm f}}{\Gamma} \left(\frac{E_{\rm r}}{E}\right)^{\frac{1}{2}} \frac{1}{1+X^2},\tag{3}$$

$$\sigma_{\rm t} = \sigma_{\rm c} + \sigma_{\rm f} + \sigma_{\rm s} \,, \tag{4}$$

where

$$X = 2(E-E_{\mathrm{r}})/\Gamma, \qquad \Gamma = \Gamma_{\mathrm{c}} + \Gamma_{\mathrm{f}} + \Gamma_{\mathrm{n}},$$

the peak height cross section $\sigma_0 = 4\pi (\lambda_r^2/4\pi^2)g\Gamma_n/\Gamma$, the potential scattering cross section $\sigma_p = 4\pi R^2$, and λ_r is the neutron wavelength at exact resonance. The cross sections are Doppler broadened by replacing $1/(1+X^2)$ and $X/(1+X^2)$ in equations (1)–(4) by the usual Voigt profiles.

The dispersion between experimental and calculated cross sections was calculated using the χ^2 goodness-of-fit statistic defined by

$$\chi^2_{N-1} \simeq \langle \sigma_{\exp} \rangle^{-1} \sum^{N} \left(\sigma_{\exp} - \sigma_{cale} \right)^2 / \sigma_{\exp} \,, \tag{5}$$

the sum being taken over all energy points.

The experimental cross sections were taken at intervals of 0.05 eV directly from the compilation of Stehn *et al.* (1965); the total cross section values were predominantly those of Pattenden and Harvey (1963) while the fission cross section used was that of Nifenecker (1964). Experimental points were averaged where necessary to obtain a single cross section value at each energy point. The data of Nifenecker were found to require a systematic energy correction in order to obtain coincidence of total and fission cross section peaks. The corrected energy E_c was found from

$$E_{\rm c} = E - \Delta E \,, \tag{6}$$

with ΔE given approximately by $0.01 E^{1.18}$. The experimental cross sections were measured at a temperature of 300°K and have not been corrected for either Doppler

| SINGLE-LEVEL RESONANCE PARAMETERS FOR ^{233}U below 11 eV | | | | | | | | | | | |
|--|---|--|------------------------------------|---|--|---|--|--|--|--|--|
| Energy (eV) | $2g\Gamma_{\rm n}$ (meV) | Γ_{γ} (meV) | $\Gamma_{\mathbf{f}}$ (meV) | Energy (eV) | $\begin{array}{c} 2g\Gamma_{\rm n} \\ ({\rm meV}) \end{array}$ | Γ_{γ} (meV) | $\Gamma_{\rm f}$ (meV) | | | | |
| $-0.590 \\ 0.325 \\ 0.645 \\ 1.455 \\ 1.807$ | $\begin{array}{c} 0 \cdot 0816 \\ 0 \cdot 0021 \\ 0 \cdot 0111 \\ 0 \cdot 112 \\ 0 \cdot 406 \end{array}$ | $71 \cdot 8 29 \cdot 2 30 \cdot 0 49 \cdot 2 69 \cdot 0$ | $1190 \\ 261 \\ 714 \\ 614 \\ 254$ | $4 \cdot 785 \\ 5 \cdot 915 \\ 6 \cdot 698 \\ 6 \cdot 825 \\ 7 \cdot 570$ | $0.335 \\ 0.175 \\ 0.473 \\ 0.743 \\ 0.0348$ | $89 \cdot 5$ 73 \cdot 7 100 \cdot 0 63 \cdot 1 59 \cdot 9 | $854 \\ 374 \\ 491 \\ 93 \cdot 7 \\ 153$ | | | | |
| $2 \cdot 302$ $3 \cdot 431$ $3 \cdot 638$ | $0 \cdot 210 \\ 0 \cdot 0775 \\ 0 \cdot 112$ | $57 \cdot 2$ $85 \cdot 4$ $93 \cdot 3$ | $55 \cdot 5$ 421 $98 \cdot 9$ | $8 \cdot 585 \\ 9 \cdot 335 \\ 10 \cdot 370$ | 0.0683 0.183 1.670 | $90 \cdot 1$ $89 \cdot 2$ $62 \cdot 4$ | 346 354 259 | | | | |



Fig. 1.—Single-level fits to (a) total cross section and (b) fission cross section for 233 U from 0 to 10 eV.

or instrumental broadening. In the energy range under consideration, the instrumental broadening is very much less than the Doppler broadening and has been neglected in calculating the cross sections.

Table 1 gives the best fitting single-level parameters below 11 eV, while Figures 1(a) and 1(b) show the calculated fits to the total and fission cross sections respectively.

TABLE 1

III. MULTILEVEL FIT

In the multilevel formulation adopted, the fission cross section in equation (3) is modified by the inclusion of the asymmetric fission interference term that approximates the interference between levels having the same spin and exit channel. Thus

$$\sigma_{\rm f}(i \to j) = \frac{8\pi (\lambda/2\pi)g(\lambda_{\rm ri}\,\lambda_{\rm rj}/4\pi^2)^{\frac{1}{2}}y_{\rm ni}\,y_{\rm nj}\,y_{\rm fi}\,y_{\rm fj}\{(E-E_{\rm ri})(E-E_{\rm rj}) + \frac{1}{4}\Gamma_i\,\Gamma_j\}}{\{(E-E_{\rm ri})^2 + \frac{1}{4}\Gamma_i^2\}\{(E-E_{\rm rj})^2 + \frac{1}{4}\Gamma_j^2\}}, \quad (7)$$

where $y_n = \pm (\frac{1}{2}\Gamma_n)^{\frac{1}{2}}$ and $y_f = \pm (\frac{1}{2}\Gamma_f)^{\frac{1}{2}}$ are proportional to the partial neutron and fission width amplitudes respectively. The interference term in the capture cross section can be ignored owing to the large number of open channels. Each channel contributes a cross term of random sign, the sum of which is distributed as a narrow Gaussian about the mean value, zero. Interference in the scattering channel may also be neglected since it is a factor of Γ_n/Γ_f ($\leq 10^{-3}$) less than the interference in the fission cross section.

TABLE 2 MULTILEVEL RESONANCE PARAMETERS FOR ²³³U BELOW 11 eV

| Energy (eV) | Γ_{n} (meV) | $\Gamma_{\gamma} \ ({ m meV})$ | Γ _f (meV) | J | Sign of $y_{\rm f}$ (equation (7)) |
|----------------|--------------------|--------------------------------|-------------------------|---|------------------------------------|
| -0.280 | 0.0466 | $37 \cdot 3$ | 1390 | | |
| 0.290 | 0.0137 | $65 \cdot 5$ | 806 | 2 | + |
| $1 \cdot 490$ | 0.141 | $34 \cdot 0$ | 508 | 2 | |
| $1 \cdot 807$ | 0.345 | $54 \cdot 1$ | 255 | 3 | |
| $2 \cdot 302$ | 0.260 | $42 \cdot 7$ | $59 \cdot 4$ | 2 | + . |
| $3 \cdot 456$ | 0.0952 | $68 \cdot 6$ | 325 | 2 | |
| $3 \cdot 613$ | 0.0854 | 70.6 | 105 | 3 | |
| $4 \cdot 620$ | 0.369 | $117 \cdot 0$ | 823 | 2 | — |
| 5.755 | 0.0669 | $124 \cdot 0$ | 159 | 3 | _ |
| $6 \cdot 840$ | 0.778 | 56.8 | 136 | 3 | + |
| 7.038 | 0.435 | $112 \cdot 0$ | 1500 | 2 | + |
| $7 \cdot 485$ | 0.0179 | $68 \cdot 8$ | 271 | 2 | + |
| $8 \cdot 700$ | 0.0228 | $108 \cdot 0$ | 172 | 3 | + |
| $9 \cdot 395$ | 0.345 | $92 \cdot 2$ | 508 | 2 | _ |
| $10 \cdot 370$ | $1 \cdot 490$ | $77 \cdot 2$ | 260 | 3 | + |

The formula ignores quantities of order Γ/D , which may lead to inaccuracies in the case of ²³³U. However, it is felt that the experimental cross sections, and particularly the fission cross section, have not been determined accurately enough to warrant a more sophisticated treatment.

Furthermore, the formula assumes a single-channel process for fission. There is considerable evidence to suggest that, at least for the states of spin and parity 2^+ , fission proceeds via more than one channel (see, for example, Wheeler 1963), and owing to cancellation the interference between levels of this spin sequence would be somewhat smaller than that predicted by equation (7). In fact, the single-channel analysis of Moore and Reich (1960) assumed that the 2^+ levels contribute a non-interfering component to the cross section. The errors arising from this



 TABLE 3

 AVERAGE PARAMETERS AND QUALITY OF FITS OBTAINED

Fig. 2.—Multilevel fits to (a) total cross section and (b) fission cross section for ²³³U from 0 to 10 eV.

assumption would be at least as large as those caused by allowing the 2^+ levels to interfere fully in one channel, since quite large fluctuations are expected even for a two-channel process.

The ease with which the above form can be Doppler broadened in terms of the standard Voigt profiles (Cook 1967) makes it particularly suited to the requirements of the reactor physicist.

Without loss of generality, all neutron width amplitudes were assumed to be positive and thus the sign of the interference term is determined by the product of signs of two fission width amplitudes. The resonance spins were determined relative to an assumed value of 3^+ for the spin of the 10.37 eV level. The two spin values and both signs for the fission width amplitude were tested for each of the levels in turn. After minimizing χ^2 with respect to the other resonance parameters, the resonance spin was assumed to be the one giving the smallest value of χ^2 . Table 2 shows the derived multilevel parameters below 11 eV with spin assignments and the relative signs of the fission width amplitudes. Table 3 shows average parameters



Fig. 3.—Interference in cross sections of 233 U showing the J values and the signs of the fission width amplitudes for resonance levels.

obtained from the derived parameters of the single-level and multilevel fits. In addition, the values of the χ^2 statistic and the calculated thermal cross sections are given as a measure of the quality of the fits obtained. Figures 2(*a*) and 2(*b*) show the calculated fits to the total and fission cross sections and Figure 3 shows the calculated fission interference term and the positions of the resonance levels.

IV. DISCUSSION

The experimental fission cross section is inadequate, but not only because an energy correction is necessary. The values given by Nifenecker (1964) in the valleys between resonances are as much as 20 barns lower than the previous data of Moore,

Miller, and Simpson (1960). For the present analysis, the more recent values were taken in all cases. Furthermore, the cross section between 8 and 10 eV is not at all well defined and this is reflected in the fact that the fit obtained is also poorer here than elsewhere. Both single-level and multilevel parameters give good fits to the cross sections, the latter being slightly superior. In common with other resonance parameter fits to the 233 U cross section, a negative energy resonance appears to be required in order to explain the large thermal cross section. In addition, a resonance is included in the single-level set of parameters at 0.645 eV to improve the fit below 1 eV. The resonance is omitted in the multilevel case.

With the exception of the narrow resonance at $2 \cdot 3 \text{ eV}$, all resonances that have been assigned to the 2⁺ spin sequence have large fission widths indicative of several open fission channels, while resonances in the other spin group are much narrower. Regier, Burgus, and Tromp (1959) have measured the ratio of symmetric to asymmetric fission from both the $1 \cdot 8$ and $2 \cdot 3 \text{ eV}$ resonances. The equality of the ratio for both levels is evidence in favour of their being of the same spin sequence, but when a fit was attempted with the spin of the $2 \cdot 3 \text{ eV}$ resonance altered, very poor agreement resulted, particularly around the resonance at $3 \cdot 6 \text{ eV}$.

There is a rather large variation to be found among the radiation widths in both cases, contrary to the expectation of constant widths when a large number of channels are open. Competition between fission and capture could cause correlations among the partial radiation widths, thus reducing the number of degrees of freedom observed in the distribution of the total radiation widths. This effect has also been noticed in 235 U (Shore and Sailor 1958; Michaudon, personal communication to compilers of B.N.L. 325, 1964).

In both single-level and multilevel fits, the average radiation width for ²³³U is found to be greater than the commonly accepted value.

V. References

- COOK, J. L. (1967).-Aust. J. Phys. 20, 67.
- FESHBACH, H., PORTER, C. E., and WEISSKOPF, V. (1954).-Phys. Rev. 96, 448.
- MOORE, M. S., MILLER, L. G., and SIMPSON, O. D. (1960).-Phys. Rev. 118, 714.
- MOORE, M. S., and REICH, G. W. (1960).-Phys. Rev. 118, 718.
- MUSGROVE, A. R. (1967).—Aust. J. Phys. 20, 73.
- NIFENECKER, H. (1964).—J. Physique 25, 877.
- NIFENECKER, H., and PERRIN, G. (1965).—Proc. Symp. on Phys. and Chem. of Fission. p. 245. (International Atomic Energy Agency: Salzburg.)
- PATTENDEN, N. J., and HARVEY, J. A. (1960).—Proc. Int. Conf. on Nuclear Structure. p. 882. (Univ. Toronto Press.)
- PATTENDEN, N. J., and HARVEY, J. A. (1963).-Nucl. Sci. Engng 17, 404.
- REGIER, R. B., BURGUS, W. H., and TROMP, R. L. (1959).-Phys. Rev. 113, 1589.
- SHORE, F. J., and SAILOR, V. L. (1958).-Phys. Rev. 112, 191.
- STEHN, J. R., GOLDBERG, M. D., WIENER-CHASMAN, R., MUGHABGHAB, S. F., MAGURNO, B. A., and MAY, V. M. (1965).—Neutron cross sections. Brookhaven National Lab. Rep. B.N.L. 325 (Suppl. No. 2, Vol. III).
- VOGT, E. (1960).—Phys. Rev. 118, 724.
- WHEELER, J. A. (1963).—"Fast Neutron Physics." (Eds. J. L. Fowler and J. B. Marion.) Vol. 2. p. 2051. (Interscience: New York.)