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INTERFERENCE EFFECTS IN FISSION CROSS-SECTIONS

PHASE ANALYSIS

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ABSTRACT

An interference analysis of the slow-neutron fission cross-sections of U^{235} and Pu^{239} was carried out. In interpreting the data, use was made of η and total cross-section measurements carried out with a pulsed-cyclotron neutron spectrometer (1, 2).

Using the cross-section-analysis method developed in (3), the authors obtained a satisfactory fit between the computed cross-section trends and the experimental results. Thanks to the comparatively high spectrometer resolution used (0.3 μsec/m) it was possible to carry out the analysis in a neutron range of up to 20 eV for Pu^{239} and 8 eV for U^{235}. Information was obtained on the relative signs of the reduced width amplitudes and the degree of interference for the strongly interfering levels. A correlation was found between the signs of the amplitudes. The number of effectively open fission channels was found to be in the region of 1 for Pu^{239} and 2 for U^{235}. Various conclusions are drawn with regard to the spins of several levels of plutonium and the relative spins of the resonances of U^{235}. Data were also obtained on the mean fission widths of levels with various spins.

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Various authors (4, 5) have drawn attention to the fact that a noticeable resonance asymmetry - absent in the case of radiative-capture cross-sections - is to be found in the slow-neutron fission cross-sections of U^{235} and Pu^{239}. This asymmetry can be accounted for in terms of interference from nearby resonance levels.

The idea of a possible interference between resonances derives from the concept of fission as a process which proceeds via a small number of open channels. By analyzing the interference effects in the fission cross-sections, it is possible to arrive at a number of conclusions with regard to the mechanism of the reaction and also to establish the number of channels that are actually open; in some cases spins can be determined and other level parameters established.

On the basis of the data hitherto available it has been possible to compare cross-sections with interference formulas only for the first resonance levels, which has detracted from the value of the information obtained. In

the case of the U^{235} cross-section analysis described in (6) and (7), for example, investigations were carried out up to a neutron energy of ~ 2.5 eV; for higher energies, qualitative results were obtained. The interference analysis of the Pu^{239} fission cross-section described in (7) only covered the first two resonance levels and even in this range the results were not absolutely certain. The analyses of the U^{233} (radiation and fission) and the Pu^{241} (total) cross-sections described in (8) and (9) cover a somewhat broader energy range.

A detailed study has now been made of the fission and radiative-capture cross-section trends with the help of data on η and the total cross-sections of U^{235} and Pu^{239} obtained with a pulsed-cyclotron neutron spectrometer (2). In this way it has been possible to carry out an interference analysis covering a broader neutron-energy range. Details of this analysis are given below together with a discussion of the results.

Method Used for Cross-Section Analysis

In attempting strictly to apply the theory of nuclear reactions to the analysis of cross-sections, it is essential to make a number of simplifications. In investigations (3) and (10) two methods of analysis are developed, on the basis of a matrix-R formalism, applicable to cases of intersecting cross-sections. We have made use of an approximation, studied in detail by Vogt in (3), which takes account of interference between a small number of levels. The number of channels effectively open is not limited and can be assessed on the basis of the results of the analysis.

The following considerations can be adduced in support of this approach. Preliminary analysis of the fission cross-sections of U^{235} and Pu^{239} show it to be possible to break down the cross-sections into a number of energy ranges in such a way that the interference between resonances in different ranges is small. It is possible to represent the contribution of neighbouring groups of levels as a slowly changing term in the cross-sections, only taking into account interferences with a small number of broad and sufficiently strong levels.

Reducing the number of levels analysed at one time considerably facilitates the calculations and has practically no effect on the accuracy of the results. At the same time this method substantially increases the reliability of the analytical data obtained.

In accordance with (3), the scattering matrix determining the energy distribution of the cross-sections can be written as follows:

$$U^J_{cc'} = e^{i(\varphi_c + \varphi_{c'})}\left[\delta_{cc'} + i \sum_{\lambda\lambda'} (\Gamma_{\lambda c})^{\frac{1}{2}} (\Gamma_{\lambda' c'})^{\frac{1}{2}} A_{\lambda\lambda'}\right] \qquad (1)$$

where $(\Gamma_{\lambda c})^{\frac{1}{2}}$ is the root of the observed partial width of transition from level λ into channel "C", with the corresponding sign (the matrix has levels of one spin only). $A_{\lambda\lambda'}$ is obtained from the equation:

$$(A^{-1})_{\lambda\lambda'} = (E_\lambda - E)\delta_{\lambda\lambda'} - \tfrac{1}{2}\, i \sum_c (\Gamma_{\lambda c})^{\frac{1}{2}} (\Gamma_{\lambda' c})^{\frac{1}{2}} \qquad (2)$$

where E is the resonance energy of level λ.

The non-diagonal components of the matrix $(A^{-1})_{\lambda\lambda'}$, which determine the interference effects in the cross-sections, can be written as follows:

$$(A^{-1})_{\lambda\lambda'} = -\frac{i}{2}\left|(\Gamma_{\lambda c})^{\frac{1}{2}} (\Gamma_{\lambda' c})^{\frac{1}{2}}\right| \cos\vartheta_{\lambda\lambda'} \qquad (3)$$

The parameter $\cos\vartheta_{\lambda\lambda'}$ - the interference phase - is linked with the number of channels participating in the reaction and with the signs of the reduced width amplitudes of the levels.

For the selection of level and interference-phase parameters, simultaneous use was made of energy-dependence data for fission and radiative-capture cross-sections and the value $\eta = \nu \cdot \frac{\sigma_f}{\sigma_a}$. The value of ν (number of secondary neutrons per fission) was assumed to be constant for all resonances (σ_f and σ_a represent fission and absorption cross-sections, respectively).

The first step was to analyse the radiation cross-sections. Because of the occurrence of a large number of transitions which are accompanied by the emission of gamma quanta, it is not possible to observe any interference effects in the total radiation cross-sections. Formally this is described by the vanishing of the non-diagonal components of the matrix $(A^{-1})_{\lambda\lambda'}$. At the same time formula (1) becomes a sum of terms similar to the Breit-Wigner single-level formula.

Where the corrections required for the Doppler broadening of the levels and spectrometer resolution are small, resonance energies and the total widths and sizes of the cross-sections in the resonance peaks can be obtained directly from a graphical analysis of the radiation cross-sections. The partial (radiation and fission) widths can then be obtained by making a

combined analysis of the fission and capture cross-sections. This procedure was adopted in dealing with the cross-sections of U^{235} in the neutron-energy range of up to 4 eV. Experimental data and an approximation of the radiative-capture cross-section in this range are shown in Fig. 1. Six resonance levels were included; account was also taken of the contribution of the level below the binding energy.

At higher neutron energies use was made of the method for obtaining level parameters first proposed in (11). Radiation widths are assumed to be constant for all levels and the fission widths are obtained from the energy dependence of η. Once the contribution of the non-interfering levels is excluded, the value of $\frac{\sigma_f}{\sigma_a}$ in the centre of the resonance can be stated, with sufficient accuracy, to be equivalent to $\frac{\Gamma_f}{\Gamma_a}$. Γ_γ was taken to be equivalent to 0.04 eV for the Pu^{239} (12) as well as for the U^{235} levels (in the case of the latter isotope, this was based on the results of our cross-section analysis at neutron energies of up to 4 eV (2)). The values for $\Gamma_{\lambda n}$ were taken from (12). The inaccuracy caused by lack of knowledge about the spin was small in the case of U^{235} ($I_o = 7/2$); in the case of Pu^{239} definite spin values were assigned to all levels included in the analysis.

In most cases the choice of the relative sign of the interference phase was made by analysing the variation of η as a function of energy. The degree of interference (the absolute value of the phase) was assessed by means of an approximate formula, valid for $(E_\lambda - E)^2 \gg (\frac{\Gamma_\lambda}{2})^2$:

$$\sigma_f \sqrt{E} = \sum_\lambda \sigma_{\lambda\tau} + \sum_{\lambda \neq \lambda'} (\sigma_{\lambda\tau}\, \sigma_{\lambda'\tau})^{\frac{1}{2}} \cos \vartheta_{\lambda\lambda'} \qquad (4)$$

where $\sigma_{\lambda\tau} = \dfrac{\sigma_{\lambda f}\, \Gamma_\lambda^2}{\mu(E_\lambda - E)^2 + \Gamma_\lambda^2}$.

The level parameters obtained were used for substitution in equations (1) and (2). After corrections had been made for Doppler broadening of the levels and for resolution, the computed results were compared with the experimental data; the parameters were corrected where necessary.

Results of Analysis

Figs. 2, 3 and 4 illustrate three resonance groups in the U^{235} fission cross-section on which interference analyses were carried out, and also the results of the approximation of the cross-sections.

The group of closely spaced levels in the region of 3 eV (Fig. 2) is the least controversial one. In the first place, only minor corrections for resolution and Doppler effect are involved and, secondly, there is no uncertainty as a result of the presence of a negative level. The energy dependences of the fission and radiative-capture cross-sections in this range also provide the clearest evidence of the importance of the interference effects. The dip in the fission cross-section curve at around 2.7 eV (Fig. 2) can only be accounted for in terms of strong "negative" interference of a level below the binding energy and with a resonance at E = 2.80 eV. The asymmetry of the resonance at E = 3.60 eV leads one to assume that there is a "positive" interference on the part of this level with the "negative" one. If the experimental points are compared with calculations which take into account the interference of these three levels, it is found that the contribution of the resonance to the cross-section at an energy of 3.13 eV is well described by a single-level Breit-Wigner formula.

Fig. 3 shows the fission cross-section of U^{235} in a neutron-energy range of 4.5 to 6.5 eV. To account for the observed energy dependence of the cross-section, it is necessary to assume maximum interference of a "negative" level with a resonance at 6.2 eV. Interference with the 5.5-eV level is less obvious; however, if this is ignored, the width that has to be assigned to the 6.2-eV level is too large to fit the cross-section in the region of 4.5 eV.

The energy range up to 2.5 eV has been investigated in detail in (6) and (7). In spite of the very great care exercised in performing the cross-section measurements in this range (12), there is considerable uncertainty about the results of the analysis because of the proximity of a "strong" negative level. Sailor (6) obtained the best fit between computed and experimental data by assuming total interference between the 0.3 and 1.15-eV levels and inserting two non-interfering levels at energies of -0.02 eV and -1.45 eV. Vogt (7) found it sufficient to introduce one interfering negative level (E = -0.95 eV). Gordeev (13) postulated one strong interfering level in the negative region and total interference of the 0.29-eV and 1.15-eV levels.

Our measurements (2) yielded results that were somewhat different from the data quoted in (12) for neutrons with energies of over 1.2 eV. Our experimental data provide a clear indication of strong interference between the 1.15-eV level and one or two "negative" levels. The 0.29-eV resonance can be reasonably described as non-interfering. Fig. 4 shows the shape of this resonance after subtraction of that part of the cross-section which is attributable to the contribution of the "negative" and 1.15-eV levels (dotted line).

The interference analysis of the cross-sections of Pu^{239} was carried out in the 7 - 24 eV energy range.

Figs. 5 and 6 show the trend of the fission cross-section for these energies. To explain the cross-section rise between resonances 10.98 and 11.95 eV (dotted line indicates symmetrical part of cross-section), one has to assume maximum interference of those two levels between one another and with a broad resonance at 15.5 eV.

The analysis of the 14.3 - 14.7 - 15.5 - 17.6 eV group of levels was more complex. The approximation shown in the diagram was obtained assuming maximum interference between resonances 14.3 - 15.5 - 17.6 eV. The strong asymmetry of the level at 17.6 eV can easily be accounted for in terms of interference with the 15.5 eV resonance. This means postulating identical spins for both levels. Direct measurement of the spins (14) yielded a value of J = 1 for the level at 17.6 eV and 0 for 14.7 eV. These two resonances cannot therefore interfere with one another. The irregularity in the energy dependence of η observed at around 14.5 eV seems to be due to interference on the part of the 14.3 and 15.5-eV levels; interference between resonances 14.3 and 15.5 eV would be insufficient to account for this. The spin of level 14.3 must therefore be equal to unity. A further point in favour of this argument is that it seems fairly improbable that two levels could be observed at a distance of less than 1/20 of the average distance for the spin in question, J = 0 (2).

The results of the Pu^{239} cross-section analysis at neutron energies up to 8 eV confirm the data obtained by Vogt (7); these are therefore not discussed in the present paper.

Table 1 sums up the data obtained from our U^{235} and Pu^{239} cross-section analyses. Data are shown for level widths, the value of $\cos \vartheta_{\lambda\lambda'}$ and the relative signs of the reduced amplitude widths $(\Gamma_{\lambda n}\ \Gamma_{\lambda f})^{\frac{1}{2}}$.

Table 1

U^{235}

λ	E eV	Γ MeV	Γ_f MeV	Γ_γ MeV	cos $\vartheta_{\lambda\lambda'}$	Relative amplitude sign	Remarks
1	-22.0	227	187			+ (conventional)	2
2	- 0.1					...	
3	0.30	135	99	36		...	1
4	1.14	150	112	38	$\cos \vartheta_{1.4} = -0.40 \pm 0.05$	-	1
5	2.04	48	10	38	$\cos \vartheta_{4.6} = -0.5 \pm 0.3$	...	1
6	2.80	200	160		$\cos \vartheta_{1.6} = 0.40 \pm 0.05$	+	1.2
7	3.13	123	79	44	$\cos \vartheta_{6.8} = -0.5 \pm 0.3$	...	1
8	3.60	83	43	40	$\cos \vartheta_{1.8} = -0.50 \pm 0.10$	-	1
9	4.84	44	4	4		...	1.2
10	5.45	63	23		$\cos \vartheta_{1.10} = 1^{-0.5}$	+	2
11	6.20	300	260		$\cos \vartheta_{1.11} = -(1^{-0.2})$	-	1.2
12	6.40	51	11		$\cos \vartheta_{10.11} = -(1^{-0.5})$	...	2
13	7.09	65	25			...	2

Pu^{239}

λ	E eV	Γ MeV	J	cos $\vartheta_{\lambda\lambda'}$	Relative amplitude sign	Remarks
1	0		1			2
2	0.30		0		...	1
3	7.84	35	1	$\cos \vartheta_{1.3} = -(1^{-0.2})$		2
4	10.9	81	1	$\cos \vartheta_{4.5} = -(1^{-0.1})$	-	2
5	11.9	23	1	$\cos \vartheta_{4.8} = -(1^{-0.2})$	+	2
6	14.3	34	1	$\cos \vartheta_{6.8} = -(1^{-0.5})$	-	2
7	14.7	23	0		...	2
8	15.5	1000	1	$\cos \vartheta_{8.9} = -(1^{-0.2})$	+ (conventional)	1.2
9	17.6	29	1		-	2
10	22.6	49	0		...	2

Remarks: 1. Graphic analysis of cross-sections
2. Constant radiation-width method

Certain remarks need to be made in connection with the level parameters quoted in Table 1. Interference analyses provide a reliable means of defining the relative signs of interference phases for strongly interacting levels. The existence of a strong "negative" U^{235} level made it possible to define relative amplitude signs not only for the closely spaced levels but also throughout the whole range of energies up to 8 eV. The amplitude signs for Pu^{239} were obtained by analysing the interference of levels with a broad resonance at 15.5 eV.

The spin assignments of the Pu^{239} levels were determined on the basis of the results of (14) and also by taking interference effects in account (negative, 0.3 eV, 14.3 eV and 15.5 eV). Since levels having different momenta cannot interfere with one another, a value of J = 1 must be assigned to the 14.3 eV, 15.5 eV and negative resonances. The value of J = 0, assigned to level 0.3 eV, was determined on the basis of the absence of interference with a negative level. While this point is not, strictly speaking, absolutely certain, the fact that maximum interference is found between all levels having an identical spin (J = 1) makes it extremely likely.

Discussion

Taking account of interference effects results in comparatively slight differences in the values of the level parameters as compared with the data obtainable with certain conventional methods. The analysis of parameter distributions, which is dealt with in (15), (16) and (17), will not therefore be discussed further in the present paper. Instead we shall attempt to analyse in greater detail the results of the phase analysis of the interference effects.

Table 2 gives data for the relative phases of cos $\vartheta_{\lambda\lambda'}$ obtained from our interference analysis of the U^{235} and Pu^{239} levels, and also various results obtained from analyses of the cross-section of U^{233} and Pu^{241} (8, 9).

The cases chosen, in our view, are such as to enable unequivocal analytical results to be obtained. Account was taken only of sufficiently close levels where mutual interference was strong. A good criterion for the selection is the value of $\frac{\Delta E_{\lambda\lambda'}}{\overline{D}}$ (ratio of the distance between interfering levels to the average distance between levels of the same spin). In the case of Pu^{239} the analysis included all pairs of levels of the same spin where $\frac{\Delta E_{\lambda\lambda'}}{\overline{D}} < 1$. In the case of the other isotopes the spins of the levels were unknown and different spin values were assigned to the non-interfering pairs.

Table 2

Isotope	E_λ, $E_{\lambda'}$	$\Delta E_{\lambda\lambda'}/\bar{D}$	$\cos \vartheta_{\lambda\lambda'}$	Remarks
Pu^{239}	10.9 -11.9	0.25	-1	Present investigation
	14.3 -15.5	0.30	-1	Present investigation
	15.5 -17.6	0.52	-1	Present investigation
U^{235}	2.8 - 3.6	0.48	-0.5	Present investigation
	5.45- 6.2	0.50	-1	Present investigation
U^{233}	1.75- 2.3	0.33	-1	(8)
	5.8 - 6.8	0.54	-1	(8)
	9.2 -10.5	0.71	-1	(8)
Pu^{241}	-0.2 - 0.3	0.19	-1	(9)
	4.5 - 6.0	0.56	+1	(9)
	6.0 -10.0	1.5	-1	(9)
	8.5 - 9.5	0.38	-1	(9)

From normal statistical considerations, it follows that $\overline{\cos \vartheta_{\lambda\lambda'}}$ must be equal to zero. It should be noted that, out of 11 pairs of resonances exhibiting a noticeable degree of mutual interference, there is only one case where the reduced amplitude signs are identical ($\cos \vartheta_{\lambda\lambda'} > 0$), i.e. where the cross-section between the resonances decreases. In the other cases, $\cos \vartheta_{\lambda\lambda'} < 0$ (the cross-section between resonances increases). One way of describing this phenomenon would be to assume, in addition to the spin, the presence of a further parameter defining phase shifts of wave functions. In this case, if levels exhibiting the same value for this parameter have Wigner-type distributions (18), then the overlap between the two systems of levels will result in the predominance of amplitudes of a different sign for the nearby levels. This effect will take place even if there is an ordered alternation in the signs of the reduced width amplitudes. The results obtained from our analysis would not be at variance with this assumption: in the case of five interfering levels of U^{235} (Table 1) there is a regular alternation of positive and negative values for $(\Gamma_{\lambda n}\ \Gamma_{\lambda f})^{\frac{1}{2}}$. Although the probability of a sequence of this sort occurring accidentally is not great (less than 10%), it is essential, in order to arrive at definitive conclusions, to study the distribution of amplitude signs for a larger number of levels.

The very fact that there is noticeable interference is indicative of the small number of open fission channels. Our interference analysis of the fission cross-section yielded a value of $\overline{|\cos \vartheta_{\lambda\lambda'}|} \approx 1$ for Pu^{239}, which corresponds to one effectively open channel. In the case of U^{235} the value obtained was $\overline{|\cos \vartheta_{\lambda\lambda'}|} \approx 0.7$ and the number of open channels is around two.

The fission-width distributions of the U^{235} and Pu^{239} levels are also indicative of a small number of channels. Furthermore, an approximation of the distributions based on Porter-Thomas curves (19) for a different number of degrees of freedom of ν yields values for ν in the region of 1 for Pu^{239} (20) and 2 for U^{235} (2).

By using the values for spins and Γ_f given in Table 1, it is possible to obtain average values of $\overline{\Gamma_f}$ for the Pu^{239} levels with spins of 1^+ and 0^+. Obviously, the values of 44 MeV for $\overline{\Gamma_f}$ $(J = 0)$ and 156 MeV for $\overline{\Gamma_f}$ $(J = 1)$ can only be considered as a first approximation; nevertheless they show that on average the fission width is greater for levels where $J = 1$.

In the case of U^{235} no data are available for determining the spins of the levels directly. It is, however, possible, on the basis of our analysis, to break down into two groups all levels up to 8 eV. The first is made up of resonances which interfere with a negative level and with one another (E_λ: -2 eV, 1.13 eV, 2.80 eV, 3.60 eV, 5.45 eV and 6.20 eV), the second includes non-interfering resonances, with E_λ equal to 0.30 eV, 2.04 eV, 3.14 eV, 4.84 eV, 6.37 eV and 7.09 eV. The level $E_\lambda \approx -0.1$ was not used in the interference analysis because of the arbitrariness of the parameter selection. The natural explanation for this breakdown is that there must be different values for J for the levels in these groups. The number of levels in each group (six and six) would also fit in with an estimate based on the expression $N_J \sim 2J+1$ (where $J = I_o \pm \frac{1}{2}$) and the value of the target-nucleus spin ($I_o = \frac{7}{2}$) for U^{235}. The average values for the fission widths of the two groups will be $\overline{\Gamma_f} = 160$ MeV and $\overline{\Gamma_f} = 37$ MeV, respectively. The values of $\overline{\Gamma_f}$ for U^{235} are different from those quoted in (2) (116 and 20 MeV). However, it should be borne in mind that the number of levels averaged out was small (six), and also that considerable uncertainty attaches to the Porter-Thomas fission-width-distribution plots contained in (2).

The small number of open channels would fit in well with the concept of the "quasi-cold" fissioning nucleus, first put forward by Bohr (21). This

investigator suggested that the probability of fission (fission width) might be strongly dependent on the magnitude of the momentum (spin) of the intermediate levels. However, in the case of Pu^{239} the observed spin-dependence proved to be at variance with that postulated by the theory.

This discrepancy can be obviated by assuming a negative parity in the case of the Pu^{239} nucleus.

The correlation occurring between signs of the reduced width amplitudes of the levels (phases) cannot be accounted for in terms of existing nuclear models. This correlation may be bound up with the important role played by single-particle states at medium excitation energies. Clearly, more experimental data and further theoretical work are needed.

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Diagrams

Fig. 1 Caption: Radiation cross-section of U^{235} up to 4 eV.
Abscissas: Neutron energy (eV).

Fig. 2 Caption: Fission cross-section of U^{235}. Neutron energies: 2.5 - 4 eV.
Abscissas: Neutron energy (eV).
Top left: barn. $eV^{\frac{1}{2}}$.

Fig. 3 Caption: Fission cross-section of U^{235}. Neutron energies: 4.5 - 6.5 eV.
Abscissas: Neutron energy (eV).
Top left: barn. $eV^{\frac{1}{2}}$.

Fig. 4 Caption: Fission cross-section and ratio $\frac{\sigma_f}{\sigma_a}$. Neutron energies: 0.03 - 1.7 eV. Results of measurements with different resolution are plotted.
Abscissas: Neutron energy.

Fig. 5 Caption: Fission cross-section of Pu^{239}. Neutron energies: 9-13 eV.
Abscissas: 10, 11 and 12 eV.

Fig. 6 Caption: Fission cross-section of Pu^{239}. Neutron energies: 13.5-18 eV.
Abscissas: Neutron energy (eV).
Top left: barn. $eV^{\frac{1}{2}}$.