SEARCH FOR INTERFERENCE BETWEEN NEUTRON RESONANCE CAPTURE AND POTEN-TIAL CAPTURE IN THE 4.9-eV RESONANCE FOR GOLD NUCLEI

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An attempt was made to observe interference between neutron resonance capture and potential capture. To this end, the capture cross sections measured during the registration of different sections of the hard part of the gamma spectrum from the reaction $Au^{197}(n, \gamma)Au^{198}$ were compared with the cross section measured during the registration of the middle part of the same spectrum. No interference was observed within the limits of experimental error. An estimate $\sigma_p < 0.5$ mb is obtained for the potential scattering cross section at the resonance energy on the assumption that the direct capture mechanism comes into play in the emission of all γ lines with energy in the interval 5.5–6.5 MeV.

INTRODUCTION

A direct mechanism may appear, alongside with the formation of a compound nucleus, when slow neutrons are captured by nuclei^[1]. Theoretical derivations with the aid of the R-matrix formalism, and considerations concerning the probability of the direct process at low energies, were presented by Lane and Lynn^[2]. Recently interest in the "direct capture" has greatly increased, since it can be used to explain some anomalies observed in the (n, γ) reaction. Many papers^[3-6] contain an analysis of the experimental material and a discussion of the results, with allowance for the possible contribution of the direct capture. However, there are no definite data as yet on the magnitude of this contribution.

It follows from a theoretical analysis^[2] that the direct capture fraction, which in general is small, should increase in those cases when the final states of the nuclei are sufficiently pure single-particle states for the radiative transitions accompanying the capture. Such cases are realized for nuclei with mass numbers in the region of 70 and below 208, which have near the ground states single-particle p-levels corresponding to a neutron on the 2p or 3p shell. Consequently, the influence of the direct capture will be manifest primarily in those nuclei, and in that part of the γ -ray spectrum corresponding to transitions to the p-levels.

Thus, by investigating the behavior of the γ -ray spectra as functions of the neutron energy, and by comparing it with the predictions of the direct-

capture theory, it is possible to obtain proof for or against the existence of direct capture. Thus, for example, evidence in favor of direct capture with emission of hard γ lines would be a correlation between the intensities of these lines and the neutron resonance widths^[2,7].

One of the manifestations of direct neutron capture should also be an interference in the partial capture cross section corresponding to one or several γ lines radiated in the direct process.

The matrix element describing the radiative capture of the neutron consists of a resonance part, containing the contributions of both mechanisms, and a nonresonant or ''potential'' part, which is due only to the direct mechanism¹⁾. The cross section will therefore display an interference between the resonant capture and the potential capture, resulting from the squaring of the sum of the corresponding amplitudes. The larger the cross section of potential capture, the more significant the interference. The corresponding calculations were made by Lovash^[8], who gives the following expression for the partial radiative-capture cross section:

$$\sigma_{i}(x) = \sigma_{i}^{\gamma} \left(\varkappa_{i} + \frac{1+2\sqrt[]{\varkappa_{i}}x}{1+x^{2}} \right), \qquad (1)$$

where σ_i^0 -cross section of the resonant capture at the resonant energy (it contains a 1/v dependence); $\kappa_i = \sigma_p / \sigma_i^0$ -ratio of the cross section of the

¹⁾The latter is correct only for an isolated resonance, for generally speaking the nonresonant part contains also "wings" of far resonances.

potential capture to the cross section of the resonant capture at resonance energy; $x = 2(E - E_0)/\Gamma$ —deviation of the neutron energy from the resonant value, divided by half the total resonance width.

The last term of this formula is due to interference; it has a common denominator with the second resonant term, and disturbs the resonance symmetry. The first term is the potential-capture cross section.

The searches for interference asymmetry in the experimental cross section curve may cast light on the existence and probability of potential capture. The chosen research object was Au¹⁹⁷ which, on the one hand, has a well isolated strong resonance at 4.91 ev and, on the other hand, has an anomalously strong group of the hardest line in the spectrum of the radiation connected with the capture. Preliminary results of the measurements were reported in ^[9]. However, as pointed out in a note added in proof to the preprint, an apparatus effect connected with the high pulse loading was mistaken for interference. In the present paper we report the results of added measurements with better apparatus.

MEASUREMENTS

The neutron source was the fast pulsed reactor of the Joint Institute for Nuclear Research, with a water moderator 36 mm thick. The half-width of the neutron pulse was approximately 50 microseconds. The investigated gold sample, measuring $100 \times 100 \times 0.5$ mm, was located 100 meters away from the reactor.

Two γ -ray detectors were used—cylindrical



NaI(Tl) crystals measuring 100×100 mm and 40×40 mm. A shield of paraffin with boron was placed between the sample and the crystals.

The γ -ray spectra were measured with a multidimensional analyzer^[10], in which the time of appearance of the pulse relative to the reactor neutron burst, and its amplitude, were recorded in arbitrary code on 15 tracks of a magnetic tape, thus providing 256 time and 128 amplitude channels. The information was then transferred from the magnetic tape to a ferrite "memory" with 2048 or 1024 channels and with readout on a printer. The dead time of the analyzer was approximately 130 microseconds. At a cycle frequency from 5 to 10 per second, it was possible to register up to 6 pulses per cycle, by using the intermediate-memory registers. The timechannel width was 64 microseconds.

In measurements with the large crystal we had to cope with undesirable apparatus effects, similar to the effect observed in ^[9]. Depending on the measurement condition, these effects varied not only in magnitude but also in the asymmetry sign. Only when the reactor power was decreased to one-tenth normal did these effects disappear. Measurements with a small crystal at normal power gave the same result. Figure 1 shows the amplitude spectra in the resonance of gold. The same figure shows the apparatus line shapes for γ -rays from a Po + Be source with 4.43 MeV energy.

The background was allowed for by making the measurements with and without the sample. An idea of the background can be gained from Fig. 2. No correction was made for the modification of

FIG. 1. Spectra of γ rays in the resonance of gold. The background has not been subtracted and amounts to approximately 2% of the total area of the spectrum. a – measurements with large detector at a reactor power of 100 W; b – measurements with small detector at 1000 W. The ordinate represents the number of counts in 8 hours (the numbers at the division should be the same as in Fig. 1a.) 1, 2, 3, 4, "control" – sections of the spectra which were used to reduce the data.



the background by the specimen because special investigations showed it to be too small.

DATA REDUCTION

Interference can be observed most reliably and simply by comparing the cross-section curve, a contribution to which from the interference term is expected, with a similar curve in which this contribution is nil or negligibly small. In our case we chose for such a control curve the cross section corresponding to the portion of the γ -spectrum which contained no lines emitted during the direct capture. To the contrary, sections containing these lines should correspond to the cross sections tested for interference. It is most convenient to take the ratios of these cross sections to the control cross section and to normalize them to unity near resonant energy.

Using (1), we can take the control cross section in the form

$$\sigma_{\text{cont}}(x) = \sigma_{\text{cont}}^{0} / (1 + x^{2}), \qquad (2)$$

and the cross section with interference in the form

$$\sigma_{\text{int}}(x) = \alpha \sigma_{\text{int}}^0 \left(\varkappa + \frac{2 \sqrt{\varkappa} x}{1+x^2} \right) + \frac{\sigma_{\text{int}}^0}{1+x^2}.$$
(3)

In writing down these formulas we assume that all the lines of the γ spectrum can be divided into two groups: lines with $\kappa_i = 0$ (the majority) and lines with $\kappa_i > 0$, which correspond to transitions to p-levels and are emitted during direct capture. In (3) α is the contribution of the lines with $\kappa_i > 0$ to the corresponding section of the apparatus spectrum, while κ is the effective ratio of the potential cross section to the resonant cross section for a mixture of these lines. FIG. 2. Comparison of the time spectra in section 2 (\bullet) and in the control section (0) of the amplitude spectrum (measurements with small detector). The first spectrum is normalized to the second at the maximum. The background in the control portion is also shown (X).

Dividing (3) by (2) and equating the ratio to unity at x = 0, we obtain

$$a(x) \equiv \frac{\sigma_{int}(x)}{\sigma_{cont}(x)} = 1 + \frac{\alpha}{1 + \alpha \varkappa} (2\sqrt[]{\varkappa} x + \varkappa x^2).$$
 (4)

The term that is linear in x insures asymmetry of a(x) relative to the point x = 0; the larger κ , the greater this asymmetry.

If we draw a smooth curve, described by formula (4), through the experimental points representing the cross section ratios, then we can determine κ . In our case (see the next section) it is more convenient to estimate κ not by this procedure, but by using the quantity

$$\Delta \equiv A_{+} - A_{-} \equiv \int_{0}^{x_{0}} a(x) dx - \int_{-x_{0}}^{0} a(x) dx = \frac{2\alpha \sqrt{\kappa}}{1 + \alpha \kappa} x_{0}^{2}$$

as a measure of the asymmetry a(x). Since $a \le 1$, we have $\Delta = 2\alpha x_0^2 \sqrt{\kappa}$ for the cases when $\kappa \ll 1$, hence

$$\varkappa = (\Delta/2\alpha x_0^2)^2. \tag{5}$$

To find the ratios a(x) it is sufficient in practice to divide the corresponding time spectra by each other, for then the factors connected with the form of the neutron spectrum and with the thickness of the specimen cancel out. Allowance for the neutron spectrometry resolution function and for the Doppler broadening cannot greatly modify the ratios a(x), and all the more so the integral quantities Δ , since the corresponding widths do not exceed the natural resonance width.

To calculate the areas A_+ and A_- it is necessary to multiply the ratios a(x) in each time channel by the energy channel width in $\Gamma/2$ units, and to sum the resultant numbers on the left and

on the right of the resonant energy within limits that are symmetrical with respect to the energy.

RESULTS AND DISCUSSION

We present the reduced measurement results (8 hours with the large detector and 38 hours with the small detector). Equal time was allotted to measurements with and without the sample.

Figure 1 shows sections of the amplitude spectra for which the ratios of the counting rates were determined as functions of the neutron time of flight. These sections were chosen in such a way that the main contribution to them was made by the hard γ lines of the spectrum. They are shown for clarity on the same figure by vertical dashed lines with heights proportional to the intensities (the data are taken from ^[11]).

The points of the time spectra lie in all cases very close to the points in the corresponding control sections (see example on Fig. 2), and accordingly none of the ratios show any noticeable asymmetry (Fig. 3). Consequently we are justified in using the present measurements only to estimate κ with the aid of formula (5). The table lists the results of the processing of all the ratios.

Proceeding to an analysis of these results, we * note first that if we assume, for example, that the



seven hardest lines of the spectrum accompany the direct capture (see Fig. 1), then, since their contribution in all the investigated portions is predominant, the interference should appear in equal fashion also in all the sections. Consequently the values of Δ listed in the table can be regarded as being more or less equivalent. Looking at these values, we can conclude that the "asymmetry" of the ratios is more likely to be due to insufficient statistics than to interference, and assume within the limits of error that Δ is equal to zero. This is also favored by the fact that when x_0 is doubled the values of Δ become even closer to zero, whereas it would follow from (5) that they should increase instead by a factor of 4.

Let us estimate the upper limit of κ . Assuming that $\Delta < 2$ and substituting in (5) the values $\alpha = 1$ and $x_0 = 60.6$, we get

$$\varkappa < 8 \cdot 10^{-8}$$
.

If we assume that the lines of this group are emitted approximately in 20% of all the capture cases^[11], and that the total capture cross section at the 4.9 eV resonance is 34,000 b^[12], then the estimate obtained above signifies that the cross section σ_p for potential capture, corresponding to the group of lines considered above, does not exceed 0.5 mb at neutron energies near 5 eV.

FIG. 3. Example of the most "asymmetrical" ratio $a(E_n)$ from among the measurements with the small detector (section 2).

Section of spec- trum	Interval of E_{γ} , MeV	A_{\pm}	A_	Δ	A+	A_	Δ
		Intervals of E _n , eV					
Large detector, 100W		$2.81 - 4,91 - 6.96; x_0 = 29,3$			$0.86-4,91-8,96; \ x_0 = 57.9$		
1 2 3	5,88-6,48 5,29-6,48 4,70-6,48	$\begin{vmatrix} 28.1 \pm 0.9 \\ 27.9 \pm 0.6 \\ 29.6 \pm 0.5 \end{vmatrix}$	${31.1 \pm 0.7 \atop 28.8 \pm 0.4 \atop 28.9 \pm 0.3}$	$\begin{array}{c} -3.0{\pm}1.1\\ -0.9{\pm}0.7\\ 0.7{\pm}0.6\end{array}$	${}^{60.7\pm2.5}_{57.3\pm1.6}_{58,2\pm1.4}$	59.0 ± 1.0 58.0 ± 0.7 57.4 ± 0.6	1.7 ± 2.7 -0.7 ± 1.8 0.8 ± 1.5
Small detector, 1000W		$3.06 - 4.91 - 6.76; x_0 = 26.4$			0.67-4,9	$91-9.15; x_0$	0 = 60.6
1 2 3 4	5,67-6.31 5,03-5,67 4,39-5.03 3,75-4.39	25.5 ± 0.4 25.4 ± 0.3 25.6 ± 0.3 $26,4 \pm 0.2$	$\begin{array}{c} 26.4 {\pm} 0.4 \\ 26.7 {\pm} 0.2 \\ 26.3 {\pm} 0.2 \\ 26.6 {\pm} 0.2 \end{array}$	$\begin{array}{c} -0,9{\pm}0,6\\ -1.3{\pm}0.4\\ -0.7{\pm}0.3\\ -0.2{\pm}0,3 \end{array}$	$\begin{array}{c} 62.1 \pm 1.3 \\ 59.6 \pm 0.9 \\ 61.1 \pm 0.8 \\ 60.3 \pm 0.7 \end{array}$	$\begin{array}{c} 61.4 \pm 0.6 \\ 61.2 \pm 0.4 \\ 60.0 \pm 0.4 \\ 60.3 \pm 0.3 \end{array}$	0.7 ± 1.5 -1.6±1.0 1.1±0.9 0.0±0.8

Values of a reas A_+ and A_- and their difference Δ .

We can likewise not exclude the possibility that our estimate of κ , and consequently of the potential-capture cross section, is not fully correct because we have "smeared out" κ over a larger number of lines than are emitted during the direct capture process. In fact, let us assume that the direct capture is realized only via emission of a single 6.25-MeV line, the intensity of which is 7%of the number of captured neutrons [11]. Simple calculations based on the known line shape of the detector show that the maximum contribution $\alpha \approx 0.3$ is made by this line in section 2 of the measurements with the small detector. Therefore we now obtain for the estimate of κ_i a value $1/\alpha^2$ = 11 times larger than before, that is, $\kappa < 9 \times 10^{-7}$, and the potential capture cross section σ_p for this line will have an upper limit of approximately 2 millibarns. For a line with a smaller yield, we would obtain a still higher estimate.

There is also some probability that our estimate of the interference is too low because of a residual apparatus effect, which exceeds the measurement error but which is cancelled out by the interference effect. This probability, however, is very low because the same should have occurred in two different measurements under different conditions.

Finally, let us consider the possible influence of the interference between resonances. Such interference in the capture cross section was first detected in the resonances of $Pt^{195[13]}$ and, as far as we know, was not observed anywhere else so far.

If we use the Breit-Wigner formula for many levels [see, for example ^[14], formula (A.20)], then we can write for the partial cross section of capture in the vicinity of the resonance, E_0 an expression which coincides exactly with (1). It takes into account the interference with one of the neighboring resonances, with parameters E'_0 , Γ'_n , and $\Gamma'_{\gamma i}$ (energy, neutron width, and partial radiative width); in this expression, however, the sign of the interference term is not known, and the role of the cross section σ_p is assumed by the wing of the neighboring resonance:

so that

$$\begin{split} \sigma_p &= \pi \lambda^2 \Gamma_n^{'} \Gamma_{\gamma i}^{'} / (E - E_0^{'})^2, \\ \varkappa_i^{'} &= \frac{\Gamma_n^{'} \Gamma_{\gamma i}^{'}}{\Gamma_n \Gamma_{\gamma i}} \frac{(\Gamma/2)^2}{(E_0 - E_0^{'})^2} \approx \left(\frac{\overline{\Gamma}}{2\overline{D}}\right)^2, \end{split}$$

where $\overline{\Gamma}$ and \overline{D} are the average total width and distance between the resonances with the same spin. The quantity κ'_i for different transitions i

and resonance $\rm E_0^\prime$ of the $\rm Au^{197}$ nucleus can assume different values on the order of 10^{-6} . Therefore the estimate $\kappa \, \sim \, 10^{-7}$ can be regarded as a limit below which the measurement of κ is practically impossible because the signs of $\sqrt{\kappa_i'}$ for different transitions i and resonances E'_0 are unknown. Some help may be rendered here by the fact that the sign of $\sqrt{\kappa'_i}$ does not depend on i for interference with potential capture. And if we gradually reduce the lower γ -ray registration energy threshold, then in the case of interference with the potential capture the observed asymmetry Δ will decrease in proportion to α , whereas in the case of interference between the resonances the decrease in Δ will be due to an increase in the number of registered transitions and their mutual cancellation in the interference term.

After this paper was submitted for publication, Wasson and Draper^[15] reported on measurements analogous to ours. They believe that they have observed interference with potential capture, and give an estimate $\sigma_p \lesssim 10 \text{ mb}$ at $E_n = 1 \text{ eV}$ (that is, 4.5 mb at $E_n = 5 \text{ eV}$). We believe that this estimate is too high and that their results imply $\sigma_p \lesssim 0.5 \text{ mb}$ at $E_n = 5 \text{ eV}$. This assumption is corroborated by the following facts:

1) Reduction of the data on Fig. 3 of the paper of Wasson and Draper^[15] by our method yields

$$\Delta = -1.2 \pm 1.6$$
 for the interval $E_n = 3.06 - 6.76$ eV,
 $\Delta = -2.5 \pm 2.1$ for the interval $E_n = 0.67 - 9.15$ eV,

which is in full agreement with our results.

2) The experimental points in ^[15] are in poor agreement with the calculated curve corresponding to $\sigma_p = 10$ mb at $E_n = 1$ eV, displaying a lower resonance asymmetry. The curve calculated from formula (4) with the interference sign reversed agrees better at $\alpha = 1$ and $\kappa = 10^{-7}$ with the points on the same figure in ^[15] than the other curves calculated by these authors.

For a final solution of the interference problem it is necessary to increase the accuracy of the measurements by several times over that in our work and in the work of Wasson and Draper^[15]. It would also be useful to investigate other suitable nuclei, particularly those in which the quantity $\overline{\Gamma}/2\overline{D}$ is even smaller.

In conclusion the authors consider it their pleasant duty to thank F. L. Shapiro for continuous interest in the work and for useful discussions, to J. Urbanec who participated during one stage of the work, G. P. Zhukov and B. E. Zhuravlev, who were' responsible for the electronic circuitry, and A. A. Loshkarev for continuous help. ¹A. V. Shut'ko and D. F. Zaretskiĭ, JETP 29, 866 (1955), Soviet Phys. JETP 2, 769 (1956).

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