Measurement of the $^{27}Al(n,\alpha)^{24}Na$ reaction cross section in the energy region 7.13 to 9.01 MeV

N. V. Kornilov, A. B. Kagalenko, and V. Ya. Baryba
Institute of Physics and Power Engineering, Obninsk, Union of Soviet Socialist Republics

S. Daróczy,* J. Csikai, Z. Papp,* and Zs. Schram
Institute of Experimental Physics, Kossuth University, Debrecen H-4026, Hungary
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The cross section of the $^{27}Al(n,\alpha)^{24}Na$ reaction relative to that of the $^{238}U(n,f)$ has been measured by the activation technique in the 7.13–9.01 MeV energy region. The neutron energy resolution varied between 70 and 130 keV (FWHM). The characteristics of the neutron source such as the mean energy of the neutrons and the energy distribution of the neutron flux have been determined experimentally and were validated with theoretical model calculations. The applied experimental techniques made it possible to decrease the overall uncertainty of the measurements to 2–3 %. The results confirm that the excitation function in the investigated energy region has a resonance structure as observed by Schmitt and Halperin in 1961. By using the data obtained for the energy distribution of the neutrons the "true" excitation function has been restored and the parameters of the Lorentz-type peaks have been calculated.

I. INTRODUCTION

The $^{27}Al(n,\alpha)$ reaction is widely used as a reference reaction both in reactor dosimetry and for measurement of the flux of monoenergetic neutrons. It has, however, been shown in Ref. 1 that the accuracy of experimental data as well as of evaluations do not yet meet the requirements set for standard reference data. All evaluations represent the $^{27}Al(n,\alpha)$ excitation function as a smooth curve. At the same time, there are experimental works which suggest that the excitation function is not a smooth function of the neutron energy. For example, in 1961 Schmitt and Halperin[2] observed several peaks in the $E_n = 6.1-8.3$ MeV energy interval at better than 100 keV energy resolution. So far, these results have not yet been supported or disproved.

On the other hand, the Advisory Group Meeting on Nuclear Standard Reference Data (Ref. 3) concluded that an improvement in the accuracy of the $^{27}Al(n,\alpha)$ cross section is only possible if experiments with 1–2 % uncertainty are performed with high neutron energy resolution.

This paper reports a measurement of the $^{27}Al(n,\alpha)$ cross section relative to the $^{238}U(n,f)$ cross section as a function of neutron energy, using the activation method. In order to fulfill the above-mentioned requirements special attention has been paid in the experiment: (a) for the detailed investigation of the neutron source so that high accuracy in the calculation of the distribution of the neutron flux along the sample could be achieved and thus an energy resolution on the order of 70 keV could be provided together with the accurate determination of the mean neutron energy; (b) and for determination of the activity of the samples and of the fission rate with 1–2 % accuracy.

II. EXPERIMENTAL

Samples of metal aluminium with diameter 19 or 10 mm and about 0.5 mm thick were mounted on the aluminium backing of the $^{238}UF_4$ layer of the fission chamber. The distance between the fissioning layer and the samples was controlled with ±0.1 mm accuracy. The chemical purity of the Al samples was better than 99.99%.

The fission chamber, continuously fed by Ar gas, contained a 0.133 mg/cm^2 thick and 19 mm diameter uranium tetrafluoride layer (99.999% ^{238}U). The layer was prepared in the Khlopin Radium Institute (Leningrad) by vacuum evaporation. Its weight and surface homogeneity were certified with better than ±1% and ±2% accuracy, respectively.

Neutrons of energy from 7.13 to 9.01 MeV were produced via the $^2H(d,n)^3He$ reaction on the tandem accelerator EGP-10M of the Institute of Physics and Power Engineering (IPPE) using a 50 mm long target cell. The deuterium gas pressure was about 26 kPa and controlled with ±0.2% accuracy. The diameter of the deuterium beam was less than 5 mm. Molybdenum foil served as the entrance window of the cell. Samples were placed 7.0±0.1 cm from the end of the gas target at the 0° direction. The maximum angle substanded by the sample was 8.7°.

Two series of irradiations have been performed. In the first one we used two samples of 19 mm diameter. In the second series we used a pair of samples with 19 and 10 mm diameter and a more uniform Mo window which provided better energy resolution.

The irradiation time varied between 3 and 8 hours. The time variation of the neutron flux was monitored and accounted for. Some irradiations were repeated to check

the reproducibility of our procedures.

The cross section of the $^{27}Al(n,\alpha)^{24}Na$ reaction has been determined by the measurement of the activity of ^{24}Na($T_{1/2}=14.96$ h, $E_\beta=1392$ keV, $I_\beta=99.92\%$, $E_{\gamma1}=1368.5$ keV, $I_{\gamma1}=100\%$, $E_{\gamma2}=2753.9$ keV, $I_{\gamma2}=99.9\%$).

III. EVALUATION OF THE EXPERIMENTAL RESULTS

A. Investigation of the parameters of the neutron source

In order to calculate the mean neutron energy and the energy resolution, the precise determination both of the thickness of the entrance window and of the deuteron energy is of primary importance.

The thickness of the Mo foil was determined on the one hand by weighting a piece of rectangular foil with known dimensions, and on the other hand, by the measurement of the ionization loss of 5.5–7.7 MeV α particles from radioactive sources. The agreement between the results of the two techniques was better than 1%. From this we concluded that the mean ionization loss of the deuteron beam in the entrance window can be calculated with the same accuracy using literature data,[4] thus providing a better than 0.1% accurate determination of the deuteron energy behind the entrance window. The α technique was also used in the determination of the inhomogeneity in the thickness of the foils, i.e., the broadening of α peaks was measured. In the first series of experiments the thickness of the entrance window was 6.45 mg/cm^2 with $\delta_R=0.46$ mg/cm^2, and in the second series it was 8.08 mg/cm^2 with $\delta_R=0.1$ mg/cm^2, where δ_R is the nonuniformity of the foil.

The deuteron energy was measured by means of the time-of-flight (TOF) method in a fixed part of the drift tube. The calibration constant c in the equation $E_d=cf^2$, where E_d is the deuteron energy and f is the NMR frequency, was determined with 0.35% accuracy at (626.7±0.5) cm flight path and at a time uncertainty of about 0.3 nsec.

The mean energy and the energy distribution of neutrons were checked in a separate transmission experiment. The (6.293±0.005) MeV resonance in the total cross section of ^{12}C was used for this purpose. For the elimination of background neutrons the TOF method has been applied. The values of the transmission were measured with ~3.5% accuracy. The experimental results were compared with Monte Carlo (MC) calculations in which the following factors were taken into account: diameter of the deuteron beam; energy loss, angular and energy straggling of deuterons in the entrance window; energy spread of deuterons caused by inhomogeneities in the Mo foil; energy loss of deuterons in the gas; reaction kinematics. In these calculations the only variable parameter was the calibration constant c.

As can be seen in Fig. 1 the agreement between the experimental and calculated results is excellent. The value of the calibration constant c thus obtained agrees within 0.2% with that measured in the TOF experiment. The above results prove that the energy distribution of the neutron flux has been calculated correctly. A more extensive description of the calculation of mean energy and energy distribution of neutrons can be found in Ref. 5.

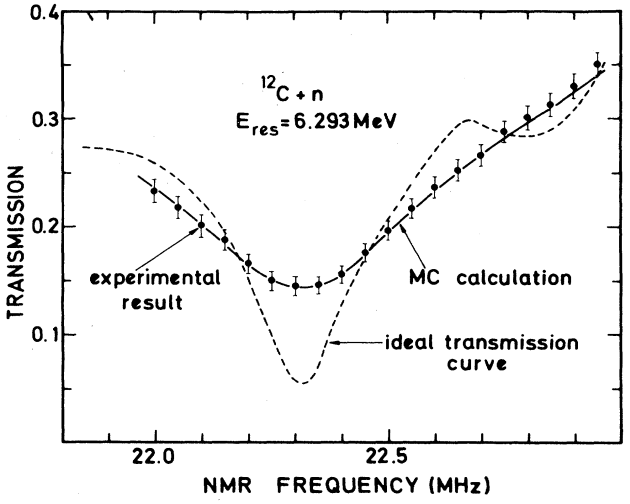

FIG. 1. Measured and calculated transmission values for neutrons on ^{12}C as a function of the NMR frequency (neutron energy). The dotted line represents the ideal transmission function, that is, if the energy distribution of neutrons is considered a δ function (the ENDF/B-V evaluation has been used for the total cross section of ^{12}C).

The MC calculation shows that in our case the energy distribution of the neutron flux is close to a normal distribution with a standard deviation, S, slowly varying with energy:

1 series (19 mm diameter): $S_1=81.48-3.68\cdot E_n$,

2A series(19 mm diameter): $S_{2A}=37.44+0.085\cdot E_n$,

2B series(10 mm diameter): $S_{2B}=40.59-1.11\cdot E_n$,

where E_n is in MeV and S obtained in keV. This energy resolution is very close to that obtainable in this kind of experiment since the resolution is limited by the interactions of the deuteron beam with the structural materials of the target assembly.

The measured neutron flux was 30–40 % lower than the calculated one, the deviation being significantly higher than the uncertainty of the MC calculation. This discrepancy is likely to be caused by local temperature rise in the gas along the beam which results in a decreasing number of deuteron nuclei available for neutron production. Therefore, it may be possible that the effective energy loss of deuterons is less than calculated, consequently, the actual mean neutron energy is ~10 keV higher than calculated. Accordingly, the accuracy of determination of the mean neutron energy in the present experiment is 15–20 keV.

B. Determination of the fission rates

Fission products have been registered in 2π geometry in a light-weight fission chamber. Losses of pulses due to the discriminator threshold have been accounted for by the extrapolation of the fragment spectrum down to zero

pulse height. This correction did not exceed 0.5%. The fragment self-absorption in the UF_4 layer was calculated by the method described in Ref. 6. For the average range of the fragments in the vacuum-evaporated UF_4 layer, $R_f=(4.76\pm0.32)$ mg/cm^2 has been used which was deduced from the experimental results of Budtz-Jørgensen and Knitter.[7] The calculated loss due to fragment self-absorption varied from 0.22% to 0.38% and was not taken into account. However, in the evaluation of the overall uncertainty of the cross-section measurements a 0.5% additional error was introduced to account for the uncertainty in the efficiency of fragment detection.

Since the count rate of the fission chamber was low (<1 cps), the influence of any electromagnetic noise on the count rate must be considered. The analysis of count rates in short time intervals of the fission chamber, current integrator and of a scintillation neutron detector makes it possible to determine the number of spurious pulses in the fission chamber. In 87% of the irradiations this correction was not greater than 1% and in 10% it was 1–2 %.

The contribution of background neutrons originated from (d,n) reactions on the structural materials has been determined in separate experiments using an empty target cell. This correction ranged from 3.5% (at E_d equivalent to $E_n=7$ MeV) to 12% (at E_d equivalent to $E_n=9$ MeV) and was measured with about 20% accuracy. For neutron energies greater than 8.5 MeV, the contribution of the $^2H(d,np)$ reaction has also been taken into account. The calculated correction was 0.5% at $E_n=8.7$ MeV and 3.1% at $E_n=9.1$ MeV. The cross section of this reaction is known to about 8% accuracy (Ref. 8) which results in less than 0.3% uncertainty in the determination of the fission rate.

C. Determination of the foil activities

Three independent methods, namely the $2\pi\beta$-γ coincidence technique, γ spectrometry and β counting in a given solid angle have been used in each case to determine the activity of the Al samples. After the irradiations the γ spectra of the foils have been measured and the other techniques were used only when the contribution of the $Al(n,p)$ reaction could be neglected. Separate experiments were made to investigate the radiochemical purity of the irradiated samples. Beta counting in the 5–150 h time interval after the irradiation showed pure ^{24}Na activity in the samples. The 14.98 h half-life obtained for ^{24}Na is in excellent agreement with the literature value (Ref. 9).

1. $2\pi\beta$-γ coincidence technique

The $2\pi\beta$-γ coincidence counting system consisted of two scintillation detectors. A thin plastic scintillator served as the β detector. For the detection of γ rays a NaI(Tl) crystal was used. In order to discriminate from the backscattered and the annihilation γ quanta, the threshold of the γ channel was set to about 600 keV. The count rates N_β, N_γ, and $N_{\beta\gamma}$ can be described by the following equations:

$$N_\beta = A\cdot[\varepsilon_\beta+(1-\varepsilon_\beta)\cdot\bar{\varepsilon}] ,$$

$$N_\gamma = A\cdot\bar{\eta} ,$$

$$N_{\beta\gamma} = A\cdot[\varepsilon_\beta\cdot\bar{\eta}+(1-\varepsilon_\beta)\cdot(\eta_1\cdot\varepsilon_2+\eta_2\cdot\varepsilon_1)] ,$$

where A is the true activity of the sample, ε_β, ε_1 and ε_2 are the efficiencies for the β particles and for the two γ lines of ^{24}Na in the β channel, η_1 and η_2 are the efficiencies for the two γ lines of ^{24}Na in the γ channel. The quantities $\bar{\varepsilon}$ and $\bar{\eta}$ are

$$\bar{\varepsilon}=\varepsilon_1+\varepsilon_2-\varepsilon_1\cdot\varepsilon_2 ,$$

$$\bar{\eta}=\eta_1+\eta_2-\eta_1\cdot\eta_2 .$$

Then

$$A=\frac{N_\beta\cdot N_\gamma}{N_{\beta\gamma}}\cdot\frac{\varepsilon_\beta\cdot\bar{\eta}+(1-\varepsilon_\beta)\cdot(\eta_1\cdot\varepsilon_2+\eta_2\cdot\varepsilon_1)}{\bar{\eta}\cdot[\varepsilon_\beta+(1-\varepsilon_\beta)\cdot\bar{\varepsilon}]}$$

$$=K\cdot\frac{N_\beta\cdot N_\gamma}{N_{\beta\gamma}} .$$

In order to determine the value of K, activity measurements have been performed with different ^{60}Co standard sources ($\varepsilon_1=\varepsilon_2$) as well as with neutron irradiated Al foils of 27–130 mg/cm^2 thicknesses ($\varepsilon_1\neq\varepsilon_2$) using 0.5, 1, and 2 mm thick plastic scintillators in the β channel. On the basis of the obtained results the value of K for Al foils with thickness ≤ 130 mg/cm^2 is $0.989<K<1$. Thus if we assume $K=1$ then the expected systematical error in our ^{24}Na activity measurements is not greater than 1.1%.

2. Gamma spectrometry

The activity of the Al foils was measured with a Ge(Li) γ spectrometer. The absolute full-energy peak efficiency of the detector for extended sources has been determined by traditional methods. The uncertainty of the efficiency curve, fitted with power functions, is about 1% in the 800–1500 keV energy region. Losses due to dead-time, random, and true pileup have been accounted for.

The results obtained by γ spectrometry are in good agreement with those deduced from the $2\pi\beta$-γ measurements. The mean activity ratio for all the measurements was

$$\langle A_\gamma / A_{2\pi\beta\text{-}\gamma}\rangle = 1.016\pm0.023 .$$

3. Beta counting

A simple end-window Geiger Muller (GM) counter taken from Debrecen to Obninsk has been used to measure the activity of the samples. In order to determine the absolute counting efficiency of the GM counter, several Al samples, identical to those used for the present cross-section measurement, were irradiated with 14 MeV neutrons and their activities were measured by a Ge(Li) γ spectrometer as well as by the GM counter in Institute of Experimental Physics (IEP). The absolute full-energy peak efficiency of this γ spectrometer was determined independently from that used in IPPE, and for the 1368.5 keV γ line of ^{24}Na it was checked with three ^{60}Co standard sources of different origin. In this way, the absolute

TABLE I. Sources of errors and their magnitudes for the calculation of the uncertainty of the $\sigma_{n,\alpha}/\sigma_{n,f}$ ratio. Φ is the neutron flux.

Source of uncertainty	Magnitude (%) $\Phi\cdot\sigma_{n,\alpha}$	$\Phi\cdot\sigma_{n,f}$
Statistical error	0.8–2	1
Number of nuclei in the samples	0.1	1
Corrections for background neutrons from (d,n) reactions on the structural materials		0.9–2.2 (E_n=7–9 MeV)
Corrections for background neutrons from the ^{2}H(d,np) reaction		<0.3
Neutron flux transfer from the UF_4 layer to the Al samples (MC calculation)	0.6	
Efficiency in fragment detection		0.5
Efficiency ratio in β detection for 10 and 19 mm diameter Al samples ($2\pi\beta$-γ method)	0.6	
Systematical error in the determination of the absolute activity of Al samples	1.1	

counting efficiency of the GM counter has been determined with 0.9% accuracy.

In the cross-section measurements, the statistical uncertainty of the β-activity measurements was better than 0.7%. The mean activity ratio for all the measurements was

$$\langle A_\beta / A_{2\pi\beta\text{-}\gamma} \rangle = 0.980 \pm 0.005 \ .$$

In the evaluation of the cross sections the activity data of the $2\pi\beta$-γ coincidence measurements were used. The results of the γ spectrometry and the β counting were considered as control data.

IV. RESULTS AND DISCUSSION

The components of the overall uncertainty of the measured $\sigma_{n,\alpha}/\sigma_{n,f}$ ratio are summarized in Table I. In the calculation of the ^{27}Al$(n,\alpha)^{24}$Na reaction cross section the data of Evaluated Nuclear Data File ENDF/B-V have been used for the ^{238}U(n,f) reaction cross section. Our results support the resonance structure already observed by Schmitt and Halperin[2] in the whole investigated energy region. On the whole, 55 cross-section measurements have been performed at different neutron energies, and from the measured data an excitation function was obtained which shows a fairly linear trend with superimposed peaks. A straight line of the form

$$\sigma_l(E_n) = 25.0 + 28.7 \cdot (E_n - 7.51) \ ,$$

where σ_l is in mb and E_n is expressed in MeV, could be fitted to the points lying between the resonances. Subtracting this linear part from the measured excitation function, the resonance structure of the cross section is more evident (see Fig. 2). Six more-or-less well-defined peaks can be seen, furthermore, where the energy resolu-

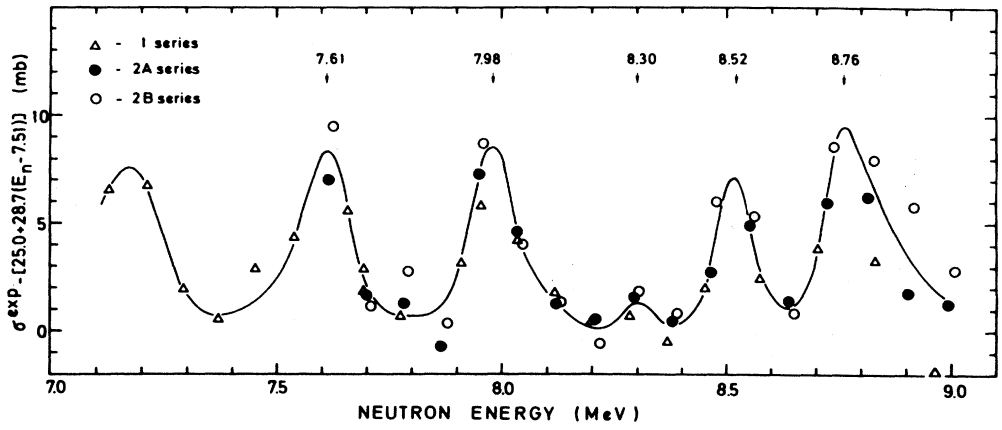

FIG. 2. The measured cross sections reduced by the appropriate $\sigma_l(E_n)$ values for the three different series of measurements. The smooth curve has been drawn by eye.

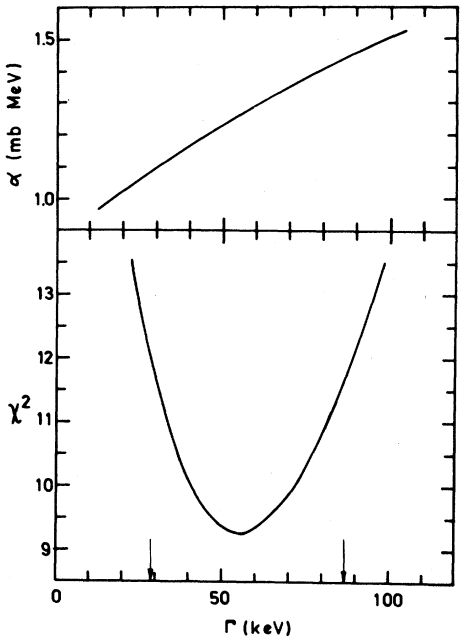

FIG. 3. χ^2 and α as a function of Γ for the resonance at 7.61 MeV. The arrows indicate the interval of the 70% confidence level.

tion is better the resonance structure is more striking.

The experimentally observable cross section is in the following relation with the "true" cross section:

$$\sigma^{\text{obs}}(E)=\int_{-\infty}^{\infty}\sigma^{\text{true}}(E')\Phi(E',E)dE' , \tag{1}$$

where $\Phi(E',E)$ is the energy distribution of the neutron flux incident on the sample and normalized to unity. Hence the "restoration" of the true cross section from the measured values is essentially the solution of an equation of the above type. The solution in the 7.37–9.01 MeV energy interval has been sought in the following form:

$$\sigma^{\text{true}}(E_n)=25.0+28.7\cdot(E_n-7.51)+\sum_{i=1}^{6}\frac{\Gamma_i}{2\pi}\cdot\frac{\alpha_i}{(E_i-E_n)^2+\Gamma_i^2/4} . \tag{2}$$

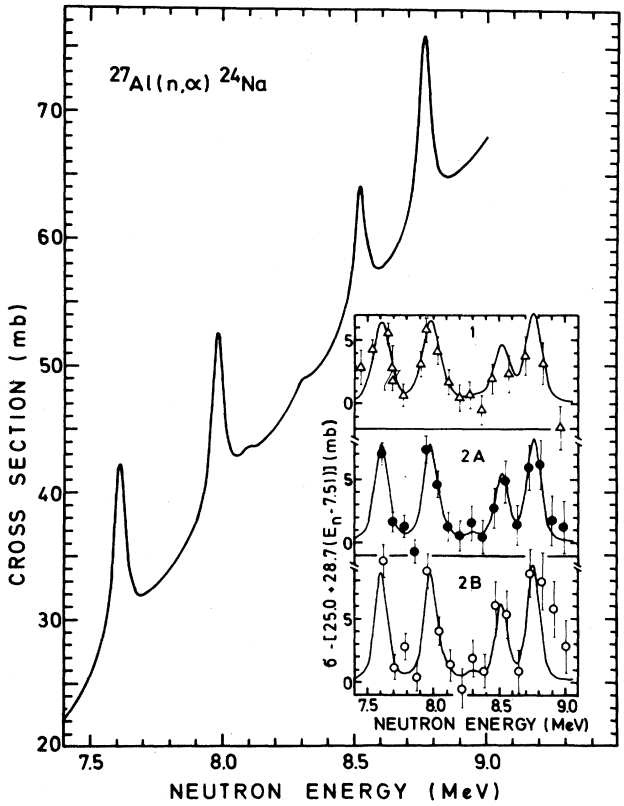

FIG. 4. The "restored" excitation function of the $^{27}Al(n,\alpha)^{24}Na$ reaction. The insert shows the resonance part of the cross section separately for the three series of measurements. The data points and the error bars denote the measured cross sections decreased by the linear part, σ_l, and the experimental errors, respectively. The smooth curves are the observable cross sections computed according to Eq. (1).

Here the peak which seems to appear at about 7.18 MeV has been omitted because evidence of it is only poorly established by the two experimental data points. On the other hand, we supposed the presence of a small peak at around 8.1 MeV to explain the shape of the resonance at about 7.98 MeV.

The $\mathbf{E}=\{E_i\}$, $\boldsymbol{\alpha}=\{\alpha_i\}$, and $\boldsymbol{\Gamma}=\{\Gamma_i\}$ parameters of this expression have been searched by iteration using the least-squares-fitting method, minimizing the following expression:

$$\chi^2(\mathbf{E},\boldsymbol{\alpha},\boldsymbol{\Gamma})=\sum_{j=1}^{3}\sum_{k=1}^{n_j}\left[\sigma_{kj}^{\text{exp}}-\int_{-\infty}^{\infty}\sigma^{\text{true}}(E',\mathbf{E},\boldsymbol{\alpha},\boldsymbol{\Gamma})\cdot\Phi(E',E_{kj},S_{kj})dE'\right]^2\Big/(\Delta\sigma_{kj}^{\text{exp}})^2 ,$$

where the energy distribution function, $\Phi(E',E_{kj},S_{kj})$, was assumed to be a Gaussian one, and E_{kj} is the mean neutron energy in the kth measurement ($k=1,2,\ldots,n_j$) of the jth series ($j=1,2,3$); σ_{kj}^{exp} is the experimentally measured cross section value at E_{kj} mean neutron energy; $\Delta\sigma_{kj}^{\text{exp}}$ is the experimental error of the σ_{kj}^{exp} result in the common 1σ sense.

Since almost every peak is fairly distinct, it was expedient to vary only the parameters of a chosen peak in every step of the iterative procedure keeping the other parameters constant. In this way, for the experimentally best resolved first two peaks the following results have been obtained:

$E_1=(7.61\pm0.01)$ MeV, $\alpha_1=(1.27\pm0.17)$ mb MeV, $\Gamma_1=(55^{+32}_{-26})$ keV ,

$E_2=(7.98\pm0.01)$ MeV, $\alpha_2=(1.21\pm0.17)$ mb MeV, $\Gamma_2=(55^{+65}_{-35})$ keV ,

TABLE II. E_i and α_i parameters of the resonances determined by a least-squares-fitting iterative procedure with the condition that $\Gamma_i = 55$ keV for all the peaks. The given uncertainties belong to the 70% confidence level. The uncertainties of the E_i values are about 0.01 MeV.

No.	E (MeV)	$\alpha \pm \Delta\alpha$ (mb MeV)
1	7.61	1.23±0.14
2	7.98	1.20±0.11
3	8.09	0.09±0.09
4	8.30	0.08±0.10
5	8.52	0.83±0.18
6	8.76	1.28±0.19

where the given uncertainties belong to the 70% confidence level. [The value of χ^2 and α as a function of Γ for the 7.61 MeV peak is demonstrated in Fig. 3. The energy parameter is fixed, the $\alpha(\Gamma)$ function was calculated from the $\partial\chi^2/\partial\alpha = 0$ equation.]

It is to be noted that the exact determination of all parameters in Eq. (2) is not possible because of the limited number of measured points. For instance, for the peak at 8.52 MeV we obtained $\Gamma = 40$ keV which is nearly the same as the former Γ values, but the corresponding error interval at the 70% confidence level was 0–150 keV. For this reason for all the peaks the value of Γ has been kept constant at 55 keV, and finally the E_i and α_i parameters have been sought under this condition. The E_i and α_i values determined in this way are given in Table II.

The "true" excitation function has been computed from Eq. (2) using the parameters obtained above and is presented in Fig. 4. The observable cross sections for the three different series of measurements have also been computed according to Eq. (1) and are demonstrated in Fig. 4 together with the $\sigma_{kj}^{\exp} - \sigma_l(E_{kj})$ and $\Delta\sigma_{kj}^{\exp}$ values separately for the three series. One can see that our "restored" excitation function is acceptably consistent with the experimental results.

The shape of our excitation function resembles that measured by Schmitt and Halperin,[2] however, their excitation function is shifted towards higher energies by 50–70 keV compared to ours. This shift is about 3 times larger than the error of the energy determination in our work. The mean separation between the peaks is about 300 keV, thus these resonances should not be energy levels of the compound nucleus, the mean separation of which is about 1 keV at about 14 MeV excitation energy. The authors mentioned above related this observation to the formation of quasimolecular states. It may be that a specific system of a ^{24}Na core and an α cluster forms when the ^{27}Al nucleus interacts with the neutron. If so, then resonances in the excitation function are observable due to the transitions between the different quasimolecular states. The mean separation of these energy levels has been estimated to be about 400 keV.[2]

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*Present address: Isotope Laboratory, Kossuth University, Debrecen H-4010, P.O. Box 8, Hungary.

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