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U. Olejniczak[1], N. A. Gundorin, L. B. Pikelner,
M. Przytuła[2], D. G. Serov

RESONANCE SPIN MEMORY IN LOW-ENERGY GAMMA-RAY SPECTRA FROM Sb, Tb, Ho AND Ta ODD-ODD COMPOUND NUCLEI

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[1] Department of Nuclear Physics, University of Lodz, Poland
[2] The College of Computer Science, Lodz, Poland

1. Introduction

In the early days of neutron spectroscopy the obtained experimental data exhibited a difference in the relative population of isomeric states from various neutron resonances in the same nuclide [1,2]. The data analysed by Draper et al. [3] allowed them to conclude that one can sometimes expect the population of the low-lying levels of the compound nucleus by cascade transitions to depend on the spin of the initial resonance state. The authors of [3] carried out an experiment designed to explain whether the differences in the low-energy gamma-ray spectra from resonances in neutron capture with an indium target were significant. The spectra in the energy region up to 350 keV from three of the ^{115}In lowest resonances: 1.46, 3.86 and 9.1 eV with spins 5, 4 and 5, respectively, were different but there was no clear correlation between the spectra and spins.

Domanic and Sailor subsequently found experimental evidence that the ratios of the populations of the isomeric 70 keV level and the ground state of ^{116}In compound nucleus for the resonance at 1.456 eV with spin 5 and for the resonance at 3.86 eV with spin 4 were quite different [4]. They concluded that difference by one in the spins of the initial resonance states significantly influenced the population ratios of the final low-lying levels. Thus one could expect that the neutron capture gamma-ray spectra for the two considered resonances should be clearly different.

Huizenga and Vandenbosch in their paper [5], which was devoted to the interpretation of isomeric cross-section ratios for radiative neutron capture, considered the question whether the value of an isomeric ratio could provide some information about the spin of an initial resonance capture state. They performed some calculations based on a simplified model of the cascade transition process assuming only dipole transitions and the application of the Statistical Model. The authors stated that the results of the calculations and the experimental data proved consistent enough to consider them as a guide to assign spins to the compound states formed in the resonance neutron capture. But they cautioned that the performed calculations were not appropriate for the cases where a statistical description was not valid.

The results of the works outlined above show that despite a large number of intermediate excited levels, which are accessible for transitions in cascade de-excitation, the memory of the spin of the initial resonance state is not lost completely, at least for some nuclei. The spin memory should be reflected in the difference between the low-energy gamma-ray spectra consisting of the transitions between the low-lying levels populated by the cascades initiated from the resonance states of different spins.

The new method of the spin assignment of s-wave neutron resonances, proposed by Wetzel and Thomas [6], is based on the conclusions presented above. The essence of the method consists in the comparison of the intensity ratios for a properly chosen pair of low-energy transitions from many resonance gamma-ray spectra. The spin memory effect manifests itself by grouping the intensity ratios around two different average values corresponding to the resonance spins $J_r = J_x \pm 1/2$, where J_x is the spin of a target nucleus. The authors of [6] tested their method on resonances with known spins and, moreover, they assigned spin values to 18 resonances of ^{167}Er and ^{187,189}Os isotopes. All the resonance states investigated in [6] belong to even-even compound nuclei.

Shortly after Wetzel and Thomas's work a number of experiments that employed the new method of spin assignment were carried out [7-11]. Therefore, apart from the investigations of the spin memory effect for those 8 isotopes studied by Wetzel and Thomas [6], that effect was also analysed in neutron resonance reactions on the target isotopes: ^{121,123}Sb [7], ^{169}Tm [8], ^{143,145}Nd [9], ^{115}In [10], and ^{175,176}Lu [11]. In one of the recent works the effect was helpful in the spin assignment for the resonances of the target isotope ^{109}Ag [12]. The prediction of the spin memory effect on the basis of a theory or a rigorous model of the cascade transition process is rather difficult and might even prove impossible. The structures of excited nuclei are very complicated and the description of the excited levels below neutron separation energy is not precisely known. The early attempts of the theoretical analysis of that process were undertaken by Pönitz [13] and Sperber [14]. The obtained results showed how complicated the problem was, and that its solution must be found for each nuclide individually. Thus one cannot tell "a priori" which pair of transitions reveals, by its intensity ratios, the existence of the spin memory effect and whether it exists at all in the nuclide in question. Nevertheless, the information on its existence and size is important not only for the method of spin assignment in the neutron spectroscopy but also for the theory of the cascade transition process.

Some time ago our attention was attracted by a remark that we found in the concluding part of the paper by Stolovy et al. [9]. The remark concerns the so called "indirect" methods of resonance spin determinations, to which Wetzel – Thomas's method belongs. The authors of [9] wrote that the methods "... should not be used in the following cases: (1) if there is a high density of low-lying states with a wide variety of spins, so that the effect of the capture state spin is diluted (most odd-odd compound nuclei);...". Despite this cautious reservation some cases of certain odd-odd compound nuclei which reveal quite considerable spin memory effect have been described. Such nuclei are, for example: two antimony isotopes [7], Thulium [8], Indium [10], Lutetium [11], and finally the Silver-109 target isotope [12]. Are they exceptions only or is it a rule?

To cast some additional light on the question we chose the appropriate isotopes of Terbium, Holmium and Tantalum for the investigation. They are all excellent candidates for further investigations into the extent to which the suspected lack of the spin memory effect in the nuclides specified above is justified. After resonance neutron capture the nuclei of the selected natural elements become the odd-odd compound nuclei. They have high density of low-lying levels, as they are middle–weight nuclei, as well as rather far from the magic ones. And as highly deformed nuclei they have many low-lying bands of collective levels with a wide variety of spins [15]. The purpose of the present work was to detect experimentally the spin memory effect in three chosen nuclides, which have extremely favourable properties for the loss of the spin memory in the cascade transition process.

2. Experimental arrangement

The investigations of the resonance radiative neutron capture were performed at the pulsed reactor IBR-30 of the Laboratory of Neutron Physics, JINR, Dubna. The use of the time-of-flight method on a 60.5 m flight path for slow neutron spectrometry allowed the resolution of neutron energy $\Delta E = 1.9 * 10^{-3} * E^{1.5}$ to be achieved, where energy is in eV. The low-energy gamma-ray spectra from the radiative capture were measured by a HPGe detector with the relative efficiency of 12% and 2.2 keV resolution at the 1332.5 keV gamma-ray ^{60}Co line.

The investigated plate samples were placed in the collimated neutron beam whose circular section was 5 cm in diameter at an angle of 45 degrees to the beam direction and at 8 cm distance from the detector head. The detector was surrounded by a shield composed of 5 cm (CH/B-10)+5 cm Pb + 1 cm Cu layers. To absorb the recycling neutrons with an energy below 0.17 eV, a cadmium sheet of 0.5 mm thickness was placed in front of the neutron collimator. The arrangement is shown schematically in Fig. 1.

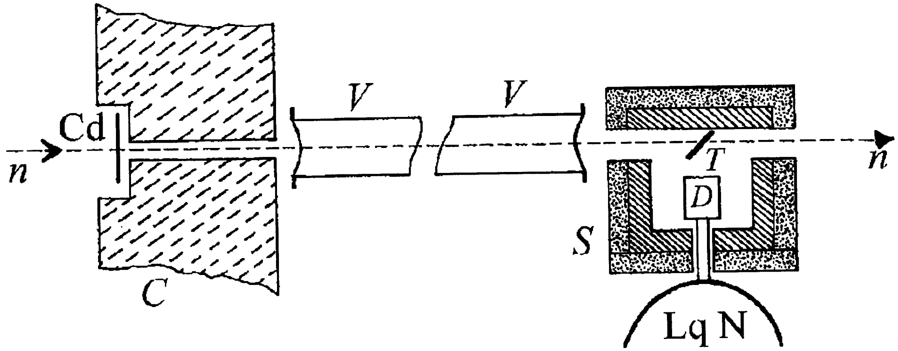

Fig. 1. Scheme of the horizontal section of the experimental arrangement:(n–n) – neutron beam, Cd – cadmium filter, C – collimator, (V – V) – vacuum beam tube, D – HPGe detector, S – detector shield, T –investigated sample, Lq N – liquid nitrogen container.

Two parameters of the detector pulses, i.e., the time-of-flight of captured neutron and pulse-height of a detected gamma-ray photon, were analysed by the computer data-acquisition system using a programme of multidimentional measurement registration DELREN, and were recorded in the memory. The obtained experimental data were located in 1024 time-of-flight and 8192 pulse-height channels. These data could be processed in the off-line procedure, which allows the selection of any required information that was accessible from the experiment.

The first measurement was treated as a test of the experimental arrangement. A natural antimony sample from Sb_2O_5 powder contained between thin aluminium sheets was used in that experiment. The spin memory effect for antimony isotopes was investigated by Bhat et al. [7], and found to be very significant.

For the subsequent measurements samples of natural Terbium, Holmium and Tantalum were used in the form of metallic plates that had thickness of 0.6 mm, 0.5 mm and 0.11 mm, respectively.

Results of measurements

A. Antimony

Natural antimony consists of two stable isotopes with mass numbers 121 and 123, and abundances 57.21% and 42.79%, respectively. Their ground state spins are $5/2^+$ for isotope 121, and $7/2^+$ for isotope 123. So , the s-wave resonances of the first isotope have spins 2^+ and 3^+, and of the second one have spins 3^+ and 4^+. The resonance energies for both isotopes and spins of a number of low-energy resonances are given in [16]. The low-energy (<511 keV) gamma-ray spectra for some resonances were measured previously by Bhat et al., and the spins of 6 resonances of ^{121}Sb were determined by the method outlined above [7].

The time-of-flight spectrum obtained from our experiment is shown in Fig. 2. The resonances of the lowest energies, i.e. 6.24 eV and 15.5 eV, belong to the A=121 isotope and have spins of $J^{\uparrow}=3^+$ and $J^{\downarrow}=2^+$, respectively. The lowest and most prominent resonances belonging to A=123 isotope are at the energies of 21.4 eV and 105.0 eV, and have spins of $J^{\uparrow}=4^+$ and $J^{\downarrow}=3^+$.

The gamma-ray spectra from the resonances at 6.24 eV and 15.5 eV are presented in Fig. 3. It is easy to notice that the intensity ratios of the neighbouring peaks corresponding to the transition energies 114.9 keV and 121.5 keV are evidently different in the spectra from the resonances of different spins.

The statistics of the counts in the spectra obtained from the resonances belonging to isotope 123 at the energies 50.3 eV and 105 eV and spins 3^+ are poor. Nevertheless, the visible difference between the spectrum from the resonance with spin 4^+ at the energy 21.4 eV and those spectra seems to show the occurrence of the spin memory effect in the odd-odd compound nucleus of the antimony-124.

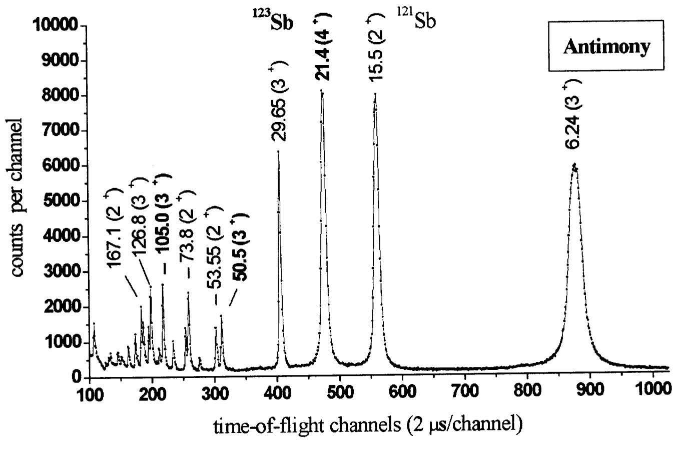

Fig. 2. Time-of-flight spectrum for the natural antimony sample. The resonances are labelled with their energies in eV and with their spins.

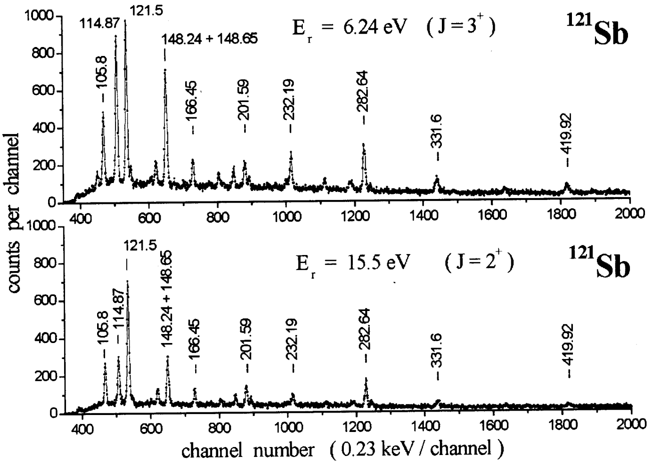

Fig. 3. The pulse-height spectra of the gamma radiation from neutron capture in the two lowest resonances in the ^{121}Sb. The spectral peaks are labelled with the corresponding transition energies in keV.

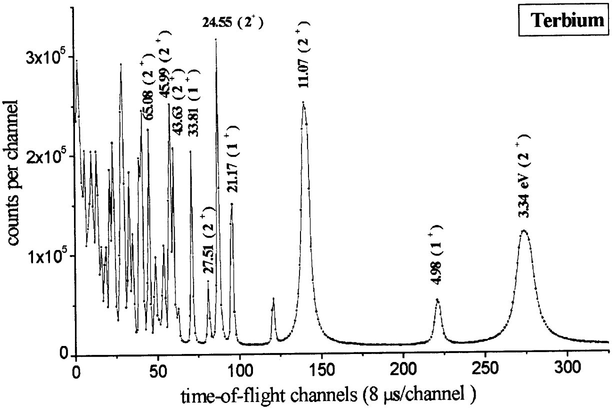

Fig. 4. The time-of-flight spectrum for the ^{159}Tb target.

B. Terbium

Natural terbium is a monoisotope of mass number 159 and ground state spin $3/2^+$, thus its neutron capture s–wave resonances can take spin values 1^+ or 2^+.

The time-of-flight spectrum obtained from a 21 hour measurement is shown in Fig. 4. In general, the resonances with spin 1^+ are visibly weaker than those with spin 2^+. For this reason only three of them could be taken into account in the analysis.

The gamma-ray spectra from the neutron capture in the resonance energies are exceptionally dense line spectra in which many peaks overlap and are therefore difficult to analyse.

C. Holmium

Natural holmium is a monoisotope of mass number 165. The ground state spin of its nuclei is $7/2^-$, and thus the spin of compound nucleus states formed after resonance neutron capture can be 4^- or 3^-.

The data for the analysis were obtained from a 20 hour measurement. In this case it was possible to gain gamma-ray spectra from 10 reasonably separate resonances, half of which had the former spin value and the other half the latter. Fig. 5 presents a summary gamma-ray spectrum covering many resonances. Some of spectral peaks are clearly separate and convenient for the analysis though some overlapping is observed, too.

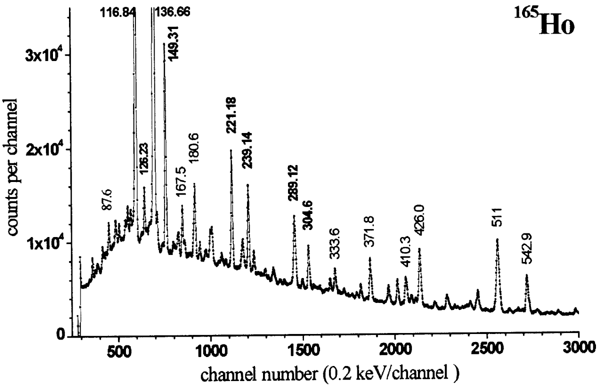

Fig. 5. The pulse-height spectrum of the gamma radiation from neutron capture in many resonances of the ^{165}Ho.

D. Tantalum

Natural tantalum consists of two isotopes with mass numbers 180 and 181. The abundance of the 180 isotope is insignificant (0.012 %) and neglected as its resonances in a measurement with a natural sample are not visible [16].

The spin of the 181 isotope ground state is $7/2^+$, and thus the spins of its s-wave resonances are 4^+ or 3^+. Fig. 6 illustrates the time-of-flight spectrum for the radiative neutron capture with the tantalum sample. The highest resonance peak on the curve corresponds to double unresolved resonances with energies 35.14 eV and 35.9 eV. They are nearly of the same strength but their spins are different.

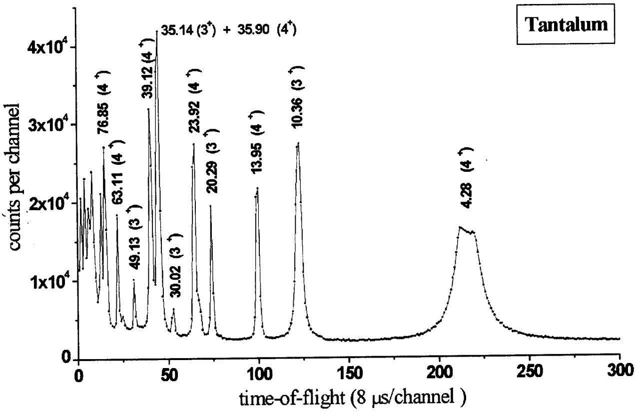

Fig. 6. The time-of-flight spectrum for the natural tantalum sample.

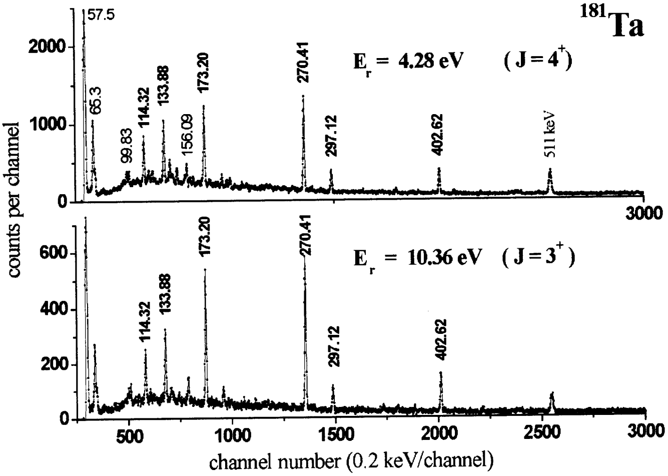

Fig. 7. The pulse-height spectra of the gamma rays from the two lowest resonances of the ^{181}Ta with different spins.

The gamma-ray spectra from two of the lowest resonances (4.28 eV and 10.36 eV) with different spins are shown in Fig. 7. These spectra look very clear with their infrequent and distinctly separate peaks. In fact, however, the spectrum from the radiative neutron capture with the tantalum sample is much more complex with hundreds of peaks corresponding to less intensive radiative transitions in the Tantalum-182 compound nucleus as demonstrated by Van den Cruyce et al. [17].

3. Analysis of experimental data

The analysis, designed to detect the existence of the spin memory effect, was conducted according to the manner presented by Wetzel and Thomas in [6]. That method allows the avoidance of the need of troublesome normalisation of the spectral data to the same resonance neutron flux, the energy dependent efficiency of the gamma-ray detector and the strength of the resonances (the “Hughes’s areas“). The idea of the method is to make comparisons of the intensity ratios for various resonances of a chosen pair of radiative transitions starting from the low-energy levels with spins J_a and J_b. In practice, instead of the transition intensities the areas under the corresponding peaks of the gamma-ray spectra, S_a and S_b, are investigated in order to obtain the ratios $R_{ab}=S_a/S_b$. In case of the occurrence of the spin memory effect the ratios R_{ab} obtained for a number of s-wave resonances are grouped around two separate average values $<R_{ab}^{\uparrow}>$ and $<R_{ab}^{\downarrow}>$ corresponding to resonances with spins $J^{\uparrow}=J_x+½$ and $J^{\downarrow}=J_x-½$ (if $J_x\neq 0$).

To quantify the strength of the effect one can accept a quantity which is the quotient of the ratios: $Q_{ab}=<R_{ab}^{\uparrow}>/<R_{ab}^{\downarrow}>$. The another measure of the effect can be expressed as the percentage of the ratio difference $|<R_{ab}^{\uparrow}>-<R_{ab}^{\downarrow}>|$ from their average value $(<R_{ab}^{\uparrow}>+<R_{ab}^{\downarrow}>)/2$, namely:

$$SME = 200\cdot[|Q_{ab}-1|/(Q_{ab}+1)]\ \%.$$

The analysis of the data was performed using many possible pairs of transitions in the studied nuclides. The searched effect manifested itself in a clear and convincing manner in a rather small part of analyzed cases. Only the most evident of them are presented below. The statistical features of R_{ab} values among resonances and their grouping around average values are illustrated graphically for those cases which are considered as convincing.

The results of the analysis of 10 resonances of the Antimony–121 target isotope are illustrated in Fig. 8a. Here the strongest spin memory effect was found for the pair of radiative transitions with the energies 114.9 keV and 121.5 keV which started from the low-energy levels of the compound nucleus with the spins $J_a=4^-$ and $J_b=1^+$. The grouping of the R_{ab} ratios into two clearly separate classes corresponding to the resonance spins 2^+ and 3^+ is evident, and their fluctuations around the respective average values for resonance spins are comprised in relatively narrow bands. The obtained value of Q_{ab}=2,57. These results confirmed those from [7] and tested successfully our experimental arrangement and the applied method of analysis.

The analysis of the data for Terbium was based on 46 combinations of transition pairs from the pulse-height spectra taken from 10 resonances. Only 3 of them have spin 1^+, and the others have spin 2^+. The obtained values of Q_{ab} are comprised between 1 and 1.92, and the highest SME=63 %. An example of a graphic illustration of the ratio grouping is shown in Fig. 8b. It corresponds to the transition pairs with the energies 193.4 keV and 158.9 keV starting from the compound nucleus levels with the energies and spins : 257.5 keV (J_a=4^-) and 222.6 keV (J_b=0^+). The spin memory effect (61 %) is unquestionable and rather strong.

In the analysis of the holmium data the gamma-ray spectra from 10 resonances were used, each half for either resonance spin. In 9 pair combinations formed from 8transitions the values of Q_{ab} that were obtained ranged from 1.18 to 1.97 with the corresponding values of SME ranging between 17% and 65%. A graphic illustration of the analysis results for the pair of transitions with the energies 149.3 keV and 239.14 keV, which started from the levels 329.8 keV (J_a=5^-) and 430 keV (J_b =2^+), is shown in Fig. 8c. The grouping of R_{ab} and their separation in the presented case are very clear.

For Tantalum the data from 7 well resolved resonances and from the peak composed of 2 unresolved resonances with different spins and the energies of 35.14 eV and 35.90 eV were analysed. Fifteen pair combinations selected from 6 transitions were taken into account. The obtained values of Q_{ab} did not exceed 1.38, and the maximum SME was 32 %. Three graphical illustrations corresponding to the highest values of Q_{ab} are presented in Fig. 9. To show the fluctuations of the individual values of R_{ab} ratios more clearly the errors are omitted. The dotted lines around the average values of $R_{ab}^{\uparrow}$ and $R_{ab}^{\downarrow}$ (solid lines) denote the standard deviation band for the fluctuating ratios in a group of a given spin value. The position of the point corresponding to the double resonance (≈ 35 eV) in the middle between two groups (in all three illustrations !) results from different spins of the unresolved resonances and their nearly equivalent “weight“ [16]. This verifies the accuracy of the measurement. The results of the analysis show a weak spin memory effect in Tantalum.

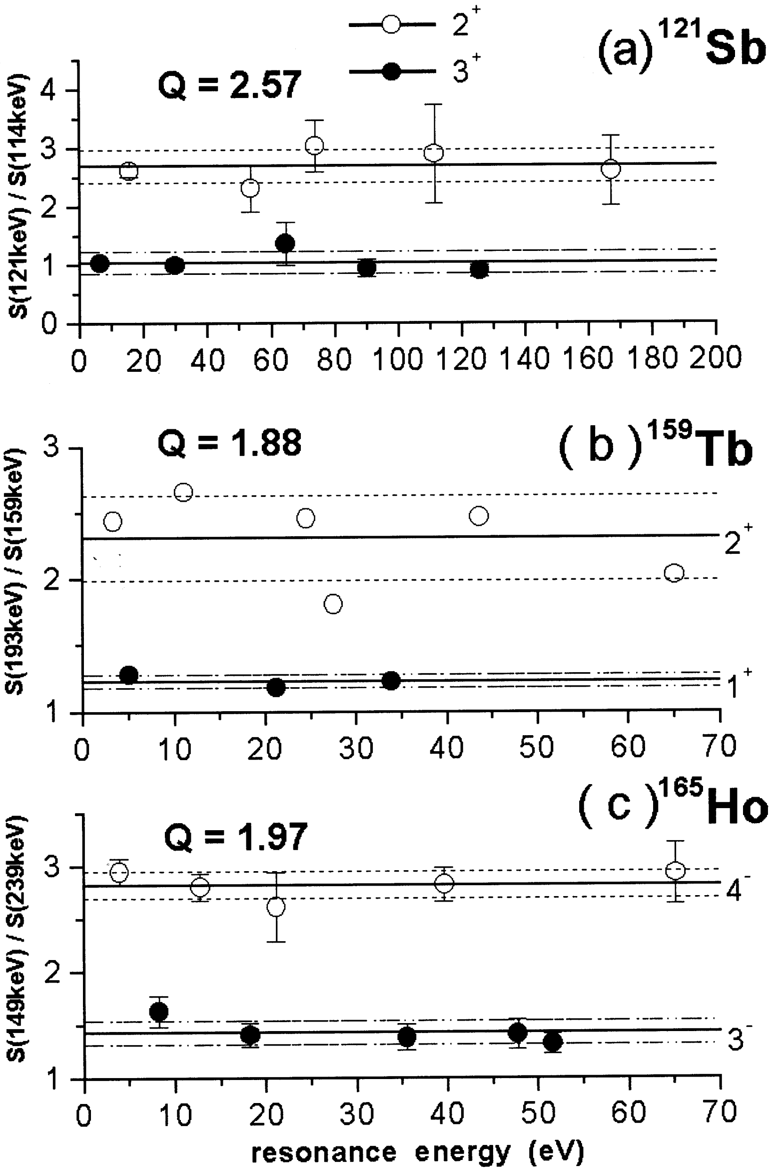

Fig. 8. (a) The ratios of the intensities of the 121.5 keV to 114.9 keV gamma rays versus the resonance energy for the ^{121}Sb target, (b) The ratios of the intensities of the transition pair with energies 193.4 keV and 158.9 keV versus the resonance energy for the ^{159}Tb, (c) The ratios of the intensities of the transition pair with energies 149.3 keV and 239.14 keV versus the resonance energy for the ^{165}Ho. The solid lines are for the average ratios of the groups corresponding to different resonance spins. The dashed lines determine the standard deviation bands for each spin group of ratios.

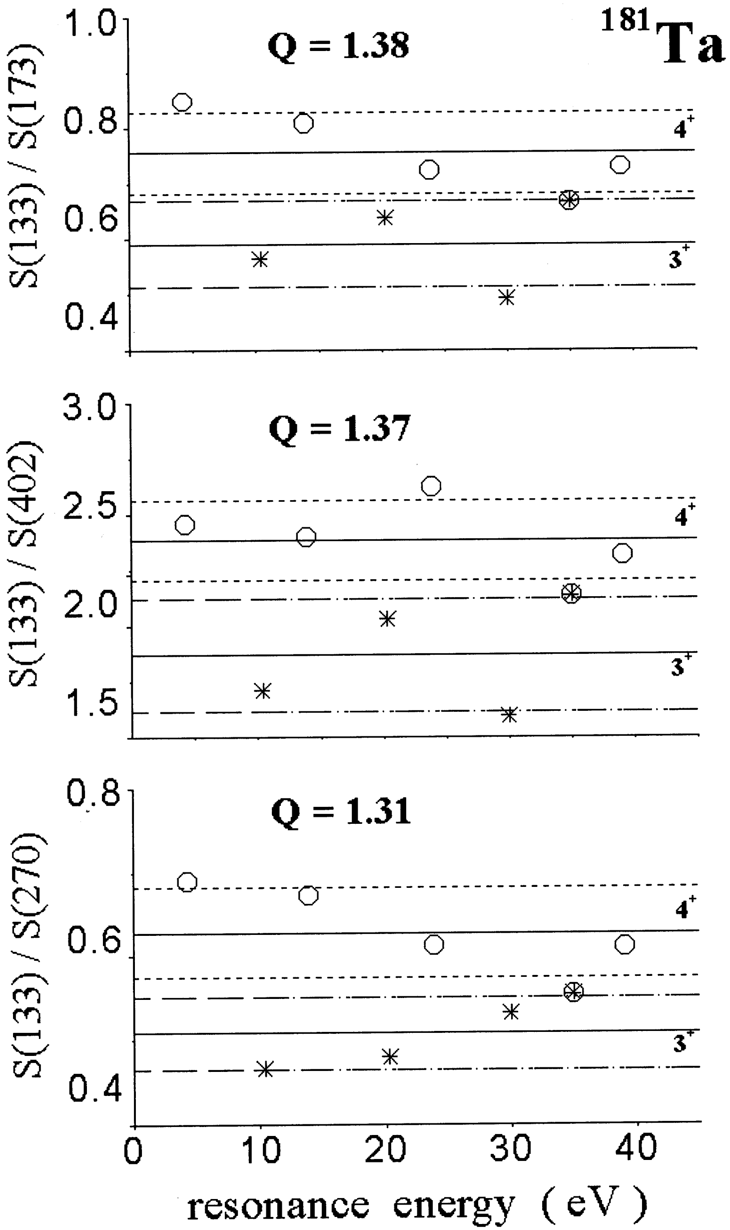

Fig. 9. The illustrations of the intensity ratio groupings for the resonances of the ^{181}Ta. The cases of the highest values of Q_{ab} are shown.

4. Remarks and Conclusions

To discuss and summarise the results let us consider those obtained in other works on spin memory effect in low-energy gamma-ray spectra and compare them for various compound nuclei. In Table I the characteristics of the spin memory effect in the odd–odd compound nuclei (including our results) and in the even–even ones are gathered. Their comparison seems to suggest that, in general, the spin memory effect in the odd–odd compound nuclei with “the high density of low-lying states with a wide variety of spins“ is practically the same as in the even–even compound nuclei situated in their proximity. An increase in the effect in the close neighbourhood of the magic number Z=50 is observed (see In, Sb, Pd, and also Ag isotope whose Z=47, which was investigated by Zanini et al. in [12]).

In the works devoted to the even–even compound nuclei [6, 9] the SME values were obtained from the ratios of pair transitions from the lowest levels of the rotational band built on the ground state. Those transitions follow each other, and if not populated from side levels, they could diminish the effect. Therefore, it cannot be excluded that the maximum effect in the even–even compound nuclei can be stronger than those observed in the works mentioned above. Some blurring of the effect can result from the manner of calculation of the areas needed for the evaluation of the R_{ab} ratios. The areas under the resonance peaks obtained from the time-of-flight spectra taken in energy windows corresponding to the transitions selected for the analysis also contain a certain contribution of some additional pulses from the recoil electrons generated in the Compton effect caused by high-energy capture photons

The presented effect could be considered as a good basis for spin assignment to neutron resonances as it was shown in works [6-12]. The measurement of the low-energy capture gamma-ray spectra for resonances using a modern germanium detector which co-operates with a time-of-flight neutron spectrometer is a relatively easy task. This method seems to be more sensitive than that one based on the “ratio of single to coincidence counts“ [18]. For example, the values of SME in the “coincidence method“ expressed in percentages are below 30% for ^{106}Pd and below 15% for ^{178}Hf compound nuclei [18], while in the “ratio of intensities method“ they are about 70% and 58%, respectively (see Table I). For ^{96}Mo compound nucleus they turned out to be the same, and equal to 25%. In the work [19], based on the former method, the obtained values of SME for the even–even compound nuclei of ^{162}Dy and ^{164}Dy do not exceed 13% while for the neighbouring even–even compound nuclei, investigated by the “ratio of intensities method”, they are greater than 30% (Table I). However, it is necessary to note that “coincidences method” uses whole gamma-ray spectrum, what means increasing of statistical precision of measurements.

The information obtained in the present work on the new nuclides which reveal the spin memory effect can be useful not only for the spin assignment method but also for the testing of the cascade transition process theory or models. The kinds of investigations, which were employed, can provide valuable data about relative populations of individual low-lying nuclear levels from de-excitation of neutron capture resonance states.

Table 1. The characteristics of the spin memory effect in compound nuclei from the resonance neutron capture

Compound nucleus			J_a , J_b	Q_{ab}	ME %	Source of information
Element	A	Z				
Odd - odd compound nuclei						
Sb	122	51	4^- , 1^+	2.57 ± 0.33	88	present work
Tb	160	65	4^- , 1^+	1.92 ± 0.28	63	
			4^- , 0^+	1.88 ± 0.27	61	
			4^- , 1^-	1.69 ± 0.24	51	
			3^+ , 0^+	1.50 ± 0.14	40	
Ho	166	67	5^- , 2^+	1.97 ± 0.19	65	
			6^+ , 2^+	1.67 ± 0.13	50	
			5^- , 3^-	1.63 ± 0.22	48	
Ta	182	73	4^+ , 1^-	1.38 ± 0.21	32	
			4^+ , 2^+	1.37 ± 0.19	31	
			4^+ , 2^-	1.31 ± 0.13	27	
In	116	49	5 , 2	2.10	71	[10]
			5 , 4	2.37	81	[10]
Sb	122	51	4^- , 1^+	~ 2.5	~ 86	[7]
Tm	170	69	3 , 0	2.11 ± 0.36	71.4	[8]
Lu	176	71	5 , 1	1.89 ± 0.04	61.6	[11]
Even – even compound nuclei						
Mo	96	42	4 , 2	1.28 ± 0.08	24.6	[6]
Pd	106	46	4 , 2	2.06 ± 0.09	69.3	[6]
Ba	136	56	4 , 2	1.52 ± 0.24	41.3	[6]
Nd	144	60	4 , 2	1.46 ± 0.04	37.4	[9]
Nd	146	60	4 , 2	1.38 ± 0.04	31.9	[9]
Er	168	68	6 , 4	1.75 ± 0.06	54.5	[6]
Hf	178	72	6 , 4	1.82 ± 0.09	58.1	[6]
W	184	74	4 , 2	2.06 ± 0.43	69.3	[6]
Os	188	76	4 , 2	1.93 ± 0.60	63.5	[6]
Os	190	76	4 , 2	1.73 ± 0.04	53.5	[6]

For this reason the results for Nd isotopes would be probably higher than those given in [9].

Our results confirm the previously observed occurrence of the spin memory effect in the odd-odd compound nuclei. The analysis done in this work shows that the observed strength of the effect depends on the selected pair of transitions starting from certain low-energy levels that differ much in their spins, J_a and J_b.

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Запоминание спинов резонансов
в спектрах низкоэнергетических гамма-лучей
нечетно-нечетных компаунд-ядер Sb, Tb, Ho и Ta

С помощью детектора из сверхчистого германия на импульсном реакторе ИБР-30 (ОИЯИ, Дубна) измерены спектры низкоэнергетических гамма-лучей резонансного захвата нейтронов на естественных смесях изотопов Sb, Tb, Ho и Ta. Эффект запоминания спинов резонансов в спектрах нечетно-нечетных компаунд-ядер ^{122}Sb, ^{160}Tb и ^{166}Ho проявился отчетливо. Для компаунд-ядра ^{182}Ta он оказался слабым.

Работа выполнена в Лаборатории нейтронной физики им. И. М. Франка ОИЯИ.

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Resonance Spin Memory in Low-Energy Gamma-Ray Spectra
from Sb, Tb, Ho and Ta Odd-Odd Compound Nuclei

The low-energy gamma-ray spectra from neutron resonance capture with natural samples of Sb, Tb, Ho and Ta were measured using HPGe detector at IBR-30 pulsed reactor (JINR, Dubna). The resonance spin memory effect in the spectra from the odd-odd compound nuclei of ^{122}Sb, ^{160}Tb and ^{166}Ho was found to be quite distinct. For the ^{182}Ta compound nucleus it proved to be rather weak.

The investigation has been performed at the Frank Laboratory of Neutron Physics, JINR.

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Макет *Т. Е. Попеко*

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