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FAST NEUTRON INDUCED DEPOPULATION OF THE ^{180m}Ta ISOMER

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I. Introduction

A probability of induced depopulation of isomeric states is an actual problem in physics of nuclear reactions, as well as it is important for the applications in astrophysics and induced γ-emission. The ^{180m}Ta isomer survives in nature due to the half-life $T_{1/2} \geq 10^{15}$ years, however its natural abundance is as low as 1.2×10^{-4} due to the restrictions on the production in stellar processes. The isomer is identified as a two-quasiparticle deformation-aligned configuration with $I, K^{\pi} = 9,9^{-}$ and an excitation energy $E^{*} = 75$ keV. High I and K values apply the limitations on the transitions from the isomer (spontaneous and induced) since the selection rules have to be satisfied and not many levels with the appropriate I and K quantum numbers are available in ^{180}Ta at modest E^{*}. It is worthwhile to remind that the angular momentum projection onto the symmetry axis, K, is typically well conserved quantum number in deformed nuclei at low E^{*}.

The ^{180m}Ta exotic nuclide has been used during the last decade as a target probe for the studies of the reactions with isomers. The (γ, γ') [1-5] and Coulomb excitation /6-10/ reactions were studied extensively in order to find the intermediate "activation" levels in ^{180}Ta which are responsible for the isomer depopulation to the ground state, ^{180g}Ta ($T_{1/2} = 8.15$ h). The activation levels should possess the K-violating wave function because their decay to the g.s. band requires a strong change of K. So, their properties are somewhat special and interesting for the detailed study.

At the original series of experiments [1, 2] the high enough yield of the ^{180g}Ta activation was observed at the bremsstrahlung end-point energies in the range $E_e = (2.5 - 5.0)$ MeV. The absolute probability of depopulation was deduced recently [4] after the experiment performed at $E_e = (5.4 - 7.6)$ MeV with application of the yield normalization by the ^{232}Th (γ, f) reaction. The low-energy range (1.0 - 2.5) MeV was covered in the last-year high-sensitivity experiment [5] performed at Stuttgart using the 150 mg, 5% enriched ^{180m}Ta target.

The Coulomb excitation results [6-9], obviously, support the presence of weak activation level at the excitation energy near 1 MeV in ^{180}Ta. More intensive K-mixing (K - violating) states are known from the (γ, γ') measurements, refs. [1, 2, 5]. In ref. [4] it was shown that the depopulation probability is strongly increased with the photon energy. Thus, weak and incomplete K-mixing at low and moderate excitations is changed to an almost complete K-mixing at E^{*} above 6-7 MeV. This conclusion is, obviously, in correlation with the results of K-hindrance factors systematics [11], demonstrating the strong decrease of reduced hindrance factors with the excitation energy of an isomeric state above the rotational energy.

In such a context the development of experiments on the induced isomers depopulation may be constructive for the phenomenology of the K-mixing amplitude dependent on the reaction and nucleus species and excitation energy. The neutron inelastic scattering is a new possibility which was not studied until now. The slow neutrons are obviously inefficient because of the angular momentum restrictions, $\Delta I = 8$ for the ^{180m}Ta depopulation. The residual energy after (n, n') reaction ought to be at least higher than the rotational energy in order to populate the g.s.b. at $I \geq (7-8)$. In the scattering of the 2-3 MeV neutrons a rather low probability of depopulation is expected, if one compares the residual excitation after (n, n') reactions with the photon energy in ref. [4]. However, one has to bear in mind that the inelastic scattering via compound nucleus (c. n.) formation is most important mechanism for the neutron scattering on heavy nuclei. Thus, the c. n. excitation may manifest itself in the intensive K-mixing. Then, the residual excitation plays a passive role, and the comparison with the (γ, γ′) reaction at the same excitation of the ^{180}Ta nucleus may be invalid. The situatiation can be clarified experimentally.

II. Experimental

The experiment on the searching for the ^{180m}Ta (n, n′) ^{180g}Ta reaction has been performed using the activation technique with the natural Ta target. The MeV neutrons were produced in the ^{9}Be (γ, n) reaction at the Dubna MT-25 microtron. The 7.3 MeV electron beam was transformed into the bremsstrahlung and into the neutron flux in the W and Be converters, respectively. The double conversion of the 20 μA electron beam produces more than 2 x 10^8 neutrons/s in 4π. The activated natural Ta foils together with the ^{232}Th target used as a monitor of the neutron flux were placed behind the lead brick at a distance of about 60 mm from the Be converter at 90° to the initial beam direction. The thick Cd and Ta envelopes were used as shields to suppress the thermal and resonance neutron flux. The scheme of irradiation is shown in Fig. 1. The choice of such a geometry is explained by the necessity to avoid the strong activation of Ta by slow neutrons as well as to make negligible the photon induced reaction yield. The cross-section of neutron capture by ^{181}Ta is very high at low energies, and the produced ^{182}Ta radionuclide ($T_{1/2}$ = 115 d) creates the intensive background in the induced γ-activity measurements (see below). The slow neutrons are present in the accelerator cabin, however the Cd shield absorbs completely the thermal component of the spectrum and 2 mm Ta plates work as resonant filters for the suppression of the ^{182}Ta activation. The photon flux in the location of the activated target is decreased by many orders of magnitude due to the converter-target distance, forward angle peaked bremsstrahlung yield and absorption in the shields. The yield of the ^{180m}Ta (γ, γ′) reaction was measured in experiment [4] at the location of the Ta target just behind the W converter. After that it was easy to recalculate the reaction yield to the geometry of present experiment, and it was found to be negligible in comparison with the expected (n, n′) reaction yield. Similarly, a negligible ratio of the (γ, f) to (n, f) reaction yields was found for the ^{232}Th target. The ^{181}Ta (γ, n) ^{180g}Ta reaction is completely hampered, since E_e = 7.3 MeV is below the threshold, and this background does not exist.

The expression for the neutron spectrum $N_n(E_n)$ contains the bremsstrahlung spectrum $N_\gamma(E_\gamma)$ and the excitation function $\sigma_{\gamma,n}$ of the ^{9}Be (γ, n) reaction:

$$N_n(E_n) = c \cdot N_\gamma(E_n + 1.67, E_e) \cdot \sigma_{\gamma,n}(E_n + 1.67) \cdot E_n^{-0.14}. \qquad (1)$$

The reaction threshold is 1.67 MeV and the resonance behaviour of the $\sigma_{\gamma,n}(E_\gamma)$ function is known from refs. [12, 13]. The last term in eq. (1) is the correction taking into account the spectrum softening in shields. The $N_n(E_n)$ function was calculated using the Monte-Carlo simulated $N_\gamma(E_\gamma)$ spectrum at the end-point energy E_e = 7.3 MeV, which corresponds to E_n^{max} ≈ 5.6 MeV. The $\sigma_{\gamma,n}$ and neutron spectrum $N_n(E_n)$ functions are shown in Fig. 2a. One can see the complicate form of the spectrum with maxima. The tendency of regular increase to the low E_n values is due to the bremsstrahlung spectral distribution N_γ. With this bremsstrahlung and neutron spectra only inelastic scattering and neutron capture are opened reaction channels. Thus, no intensive background activities could be produced in Ta after the irradiation, except the ^{182}Ta radionuclide.

The 10 g ^{nat}Ta target (stack of foils) was exposed during 6 h to the neutron flux in the described conditions. The target contains about 1 mg of ^{180m}Ta. The Ta foils of 50 μm thickness were distributed around the HP Ge detector for the γ-spectroscopy of the induced activity after irradiation. The self-absorption of X-rays in foil didnot exceed 10%, and the corresponding correction was introduced. The energy resolution of the detector was about 0.7 keV near E_γ = 60 keV. The activity of ^{180g}Ta ($T_{1/2}$ = 8.15 h) was searched for by the Hf K-X-ray lines, most intensive in its decay. As clear in Fig. 3, weak $K_{\alpha2}$ Hf line (54.6 keV) was fortunately detected in presence of more intensive radiation of ^{182}Ta. This line disappears after 13 h cooling, that confirms the detection of the 8 h lived ^{180g}Ta nuclide. If spectrum 3b is subtracted from 3a, one can also find the $K_{\beta1}$ Hf line (63.2 keV) but with poor statistics. As expected, ^{182}Ta was produced in a large amount due to the neutron capture on the abundant ^{181}Ta isotope. Many characteristic γ-lines as well as the W K-X-ray lines are emitted in the ^{182}Ta decay. All of them, together with the natural radioactivity lines were observed in the measured spectra. Unexpectedly, the Ta K-X-rays were also very intensive because of the fluorescence excited in the Ta foil by the β-, γ- activity of the foil itself. Other lines are in agreement with the expectation, and their affiliation is explained in Table 1. Some of them are not resolved in Fig. 3. They look as one peak, for instance, the $K_{\alpha1}$ line is typically combined with the $K_{\alpha2}$ line of the next element.

III. Results and discussion

As shown in Fig. 3, the $K_{\alpha2}$ line of Hf was observed successfully, and the statistical inaccuracy of its area was about 15%. It allows one to deduce the integral yield of the ^{180m}Ta (n, n′) ^{180g}Ta reaction and compare it with the yields of the ^{181}Ta (n, γ) ^{182}Ta and ^{232}Th (n, f) reactions detected with much better statistics in the same experiment. The fission yield in the monitoring Th target was deduced after detection of the radioactive fragments, such as ^{91}Sr, ^{92}Sr, ^{97}Zr, ^{133}I, ^{142}La, etc. The results are given in Table 2.

The experimentally determined yields can be simulated using the calculated neutron spectrum, eq. (1), and the excitation function of reaction. The σ (E_n) functions for the ^{232}Th (n, f) and ^{181}Ta (n, γ) reaction are shown in Fig. 2b in accordance with the Handbook [14]. The simulated ratio of the yields for these two reactions perfectly reproduces the measured value, the comparison is given in Table 2.

For the ^{180m}Ta (n, n′) ^{180g}Ta reaction neither cross-section nor its energy dependence are known. The threshold behaviour of the excitation function has been assumed in order to estimate the probability of the depopulation basing on the measured yield:

$$\sigma_{180g}(E_n) = \begin{cases} 0 & \text{at } E_n \le E_{th}; \\ 0.5\,\sigma_{tot} \cdot \dfrac{\sigma_g}{\sigma_g + \sigma_m} & \text{at } E_n > E_{th}, \end{cases} \qquad (2)$$

where the threshold energy is expressed:

$$E_{th} = 0.7 + \varepsilon_{kin}(n') \approx 1.3 \text{ MeV}. \qquad (3)$$

The value 0.7 MeV corresponds to the rotational energy for I = 8 in the ^{180}Ta residual nucleus. Above the threshold the cross-section is defined by the neutron absorption cross-section (= 0.5 · σ_{tot} in the black nucleus model) and by the probability of the decay to the ground state: R = $\sigma_g / (\sigma_g + \sigma_m)$. The total reaction cross-section σ_{tot} is shown in Fig. 2b in accordance with the data of ref. /14/ for the ^{181}Ta nucleus. It is impossible to expect any significant difference in σ_{tot} for ^{180m}Ta and ^{181}Ta nuclei, since it's defined by the geometry parameters in the MeV range. The mean kinetic energy of the scattered neutron ε_{kin} (n$'$) was calculated assuming the mechanism of statistical evaporation from the c. n. The mean c. n. excitation is defined by the mean projectile energy $\overline{E}_n$ and the latter one was calculated as follows:

$$\overline{E}_n = \int_0^{E_n^{max}} E_n\,\sigma(E_n)\,N_n(E_n)\,dE_n \Big/ \int_0^{E_n^{max}} \sigma(E_n)\,N_n(E_n)\,dE_n . \qquad (4)$$

The numerical values are given in Table 2.

Eqs (1) - (3) were used to simulate the yield of the ^{180m}Ta (n, n$'$) ^{180g}Ta reaction, varying the parameter R for the fit. It was found that the value R $\approx$ 0.4 is necessary to reproduce the measured yield of this reaction. It means that the probability is as high as 40% for the isomer depopulation to the g. s. after (n, n$'$) reaction. Such a high probability was not expected taking into account the results of ref. [4] on the (γ, γ') - reaction for the same excitation energy of the ^{180}Ta nucleus.

This fact can be explained only assuming that the specific K-configuration of the isomer is melted immediately after the neutron absorption because of high enough excitation of the compound nucleus (above 9 MeV). The following emission of the neutron and γ-rays is governed by the standard statistical laws. Accordingly to them, the residual excitation should definitely exceed the rotational energy (eq. (3)), all transitions should satisfy the selection rules by spin and parity, and the position of residual nucleus in the (E^*, I) coordinates is defined by the level density and not by the initial quasiparticle configuration. In the photon scattering the nucleus is not going through the stage of highly-excited compound nucleus, and this influences the final probability of the ^{180m}Ta depopulation making it different in two reactions at the same residual excitation.

Accepting this explanation, the value of $R = \sigma_g/(\sigma_g+\sigma_m) = 0.4$ determined from the (n, n$'$) reaction experiment is placed in Fig.4 at the position of mean c. n. excitation, not mean excitation of the residual nucleus for comparison with (γ, γ') reaction data [4]. One can see that plotting the (n, n$'$) result in this way prevents a strong disagreement between the different reaction studies which would arise if the (n, n$'$) datum is placed according to the residual excitation of ^{180}Ta. The most appropriate comparison between these reactions is therefore obtained by considering the energies of different nuclei, ^{180}Ta for (γ, γ') and ^{181}Ta for (n, n$'$), reached during the scattering processes, but at equivalent reaction stages. The inaccuracy of the measured depopulation probability R includes statistical error (15%) and 10% error due to the normalization and simulation procedures. The horizontal bars show the estimated half-width of the E^* distribution due to the continuous neutron spectrum. It would be, of course, important to repeat this experiment with the quasimonochromatic neutron spectrum. Until then it is difficult to comment upon the energy dependence of the cross-section, although its threshold behaviour seems to specific to the ^{180m}Ta (n, n$'$) ^{180g}Ta reaction. In any event, almost complete K-mixing is found in the deformed nucleus at $E^* \geq B_n$.

In both reactions after photon absorption and after neutron scattering the intermediate nucleus has a few MeV of excitation energy and high enough spin I $\approx$ 8. There is no restriction for the decrease of E^* and I in the stretched γ- cascades by the rotational bands linked to the g. s. band. Why it happens with high probability after (n, n$'$) reaction and with much lower probability after photon absorption? That is because the groups of the populated states are not identical in both reactions. The photon absorption onto K = 9 state populates mostly states with high K and quite a few K-mixing states available at $E^* \approx$ 2-3 MeV. The stretched cascades with a regularly decreasing K are unknown according to the modern nuclear spectroscopy data. Thus, high K states are decayed to the initial K = 9 isomeric state. The neutron emission from the highly excited c. n. populates all levels, mainly with low K values, because K-quantum number is mostly nonconserved for the states at $E^* > 9$ MeV. After neutron emission γ- cascades follow to the g. s. This is an explanation based on the K-mixing at high E^*.

The only alternative would to attempt to explain the large ΔI and ΔK values observed by the angular momenta carrying in (out) by the incident (evaporated) neutron. To evaluate this possibility, the spin distribution of the ^{180m}Ta (n, n$'$) reaction product was calculated for input (output) kinetic energies of the neutron of 3.0 (1.5) MeV and for the residual excitation energy 1.575 MeV, respectively (to account for the isomeric energy release). The rules of vector addition of the angular momenta, the transmission coefficients T_l for fast neutrons and the spin dependence of the level density are the most important ingredients of these calculations. The kinetic energies chosen for in (out) neutrons in the calculation are higher than the mean values given above for the actual experiment discussed here, thus the role of the neutron angular momenta is overestimated in this test calculation. In the deduced distribution shown in Fig. 5, one can see that only 12% corresponds to the low-spin part of the distribution with $l \leq 4$. This is not sufficient to explain the 40% probability for the reaction branch ending up the 1^+ g. s. in ^{180}Ta. The value ΔK = 3 can be released with a reasonable probability in a cascade of 2 - 3 statistical γ- quanta. However, ΔK = 4 and 5 are improbable for a random statistical cascade, and one returns again to the idea of a cascade with a regular decrease of the K quantum number. Since this idea has no support in nuclear spectroscopy, one has to conclude that the angular momenta of the neutrons and gammas cannot account for a large part of the release of ΔK = 8 in the (n, n$'\gamma$) - reaction. And the only explanation is the K-mixing properties of the intermediate compound-nucleus states.

IV. Conclusion

The probability of the ^{180m}Ta isomer depopulation to the ground state by an neutron inelastic scattering reaction was measured for the first time. The results show definitively that the specific K-configuration of the isomer does not survive after the neutron absorption, and the following emission of the neutron and γ-rays is governed by the statistical laws being restricted mostly by energy and angular momentum conservation, and not by structure selectivity.

V. Acknowledgments

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Table 1.

X- and γ-ray lines of activity induced in Ta by neutron irradiation

E_γ (keV)	Affiliation
53.2	nat. backgr.
54.6	$K_{\alpha 2}$ Hf (^{180}Ta)
55.8 56.3	$K_{\alpha 1}$ Hf $K_{\alpha 2}$ Ta
57.5 58.0	$K_{\alpha 1}$ Ta $K_{\alpha 2}$ W (^{182}Ta)
59.3	$K_{\alpha 1}$ W (^{182}Ta)
63.2 63.3	$K_{\beta 1}$ Hf (^{180}Ta) nat. backgr.
65.0 65.2 65.7	$K_{\beta 2}$ Hf $K_{\beta 1}$ Ta γ ^{182}Ta
67.0 67.2 67.7	$K_{\beta 2}$ Ta $K_{\beta 1}$ W (^{182}Ta) γ ^{182}Ta
69.1	$K_{\beta 2}$ W (^{182}Ta)

Table 2.

Yields of the fast neutron induced reactions

Reaction	B_n (MeV)	Mean E_n (MeV)	C. n. mean E* (MeV)	Normalized yield	
				exp.	simulation
^{232}Th (n, f)	4.79	2.7	7.5	1	1
^{181}Ta (n, γ) ^{182}Ta	6.06	0.42	6.5	6.2 ± 0.2	6.18
^{180m}Ta (n, n$'$)^{180g}Ta	7.65	2.3	9.9	13.4 ± 3.8	13.4*)

*) Using $\sigma_g/(\sigma_g + \sigma_m) = 0.403$

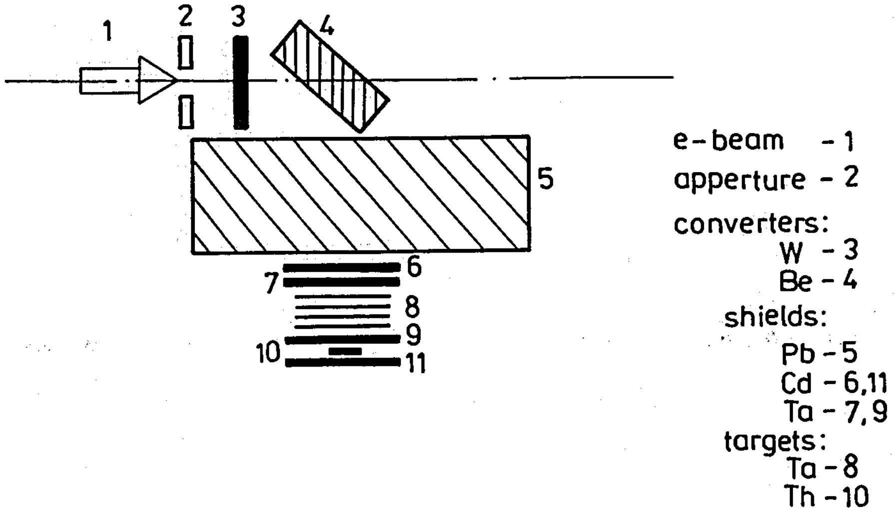

Fig. 1. Scheme of the experiment on activation of Ta by fast neutrons.

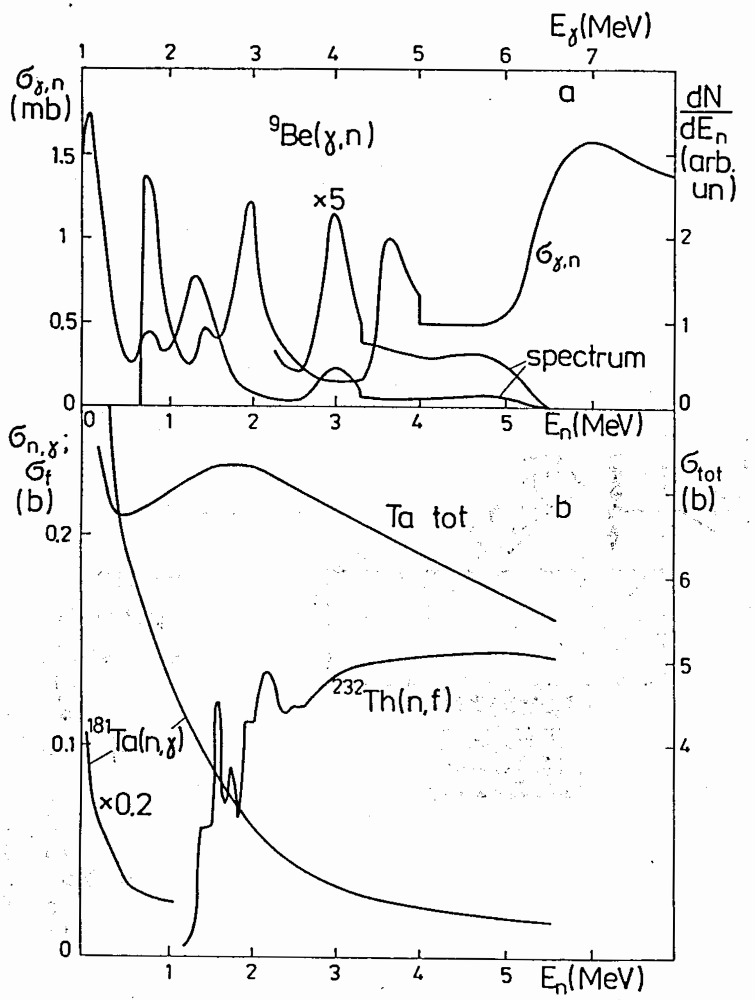

Fig.2. a) Cross-section of the ^{9}Be (γ, n) reaction as a function of E_γ [12, 13] and the spectrum of neutrons produced in this reaction by the bremsstrahlung with E_e = 7.3 MeV.

b) Energy dependencies of the cross-sections of neutron-induced reactions: ^{232}Th (n, f), ^{181}Ta (n, γ) and total reaction cross-section for ^{181}Ta as given in ref. [14].

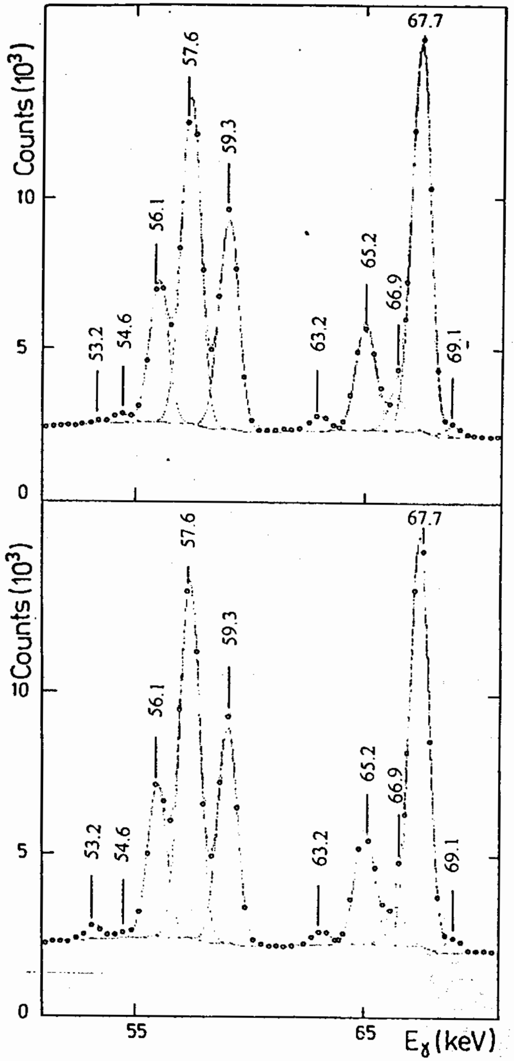

Fig. 3. Gamma-ray spectra of the activated by fast neutrons Ta sample measured after the cooling during 2 h (a) and 13 h (b). The storage time was about 11 h in both cases.

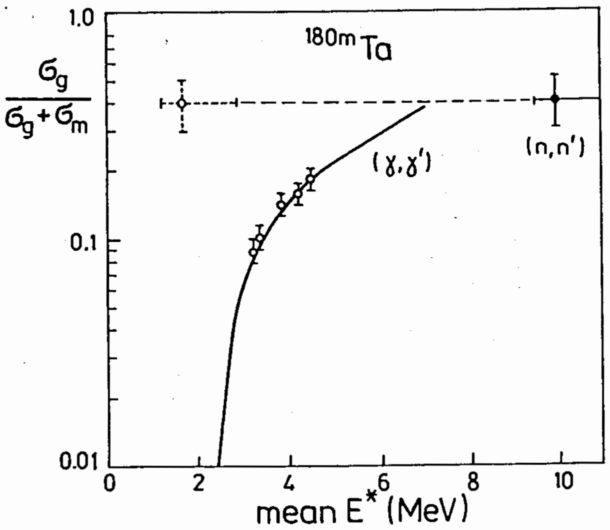

Fig. 4. Probability of the ^{180m}Ta isomer depopulation measured in the (γ, γ$'$) and (n, n$'$) reactions as a function of the compound nucleus excitation energy.

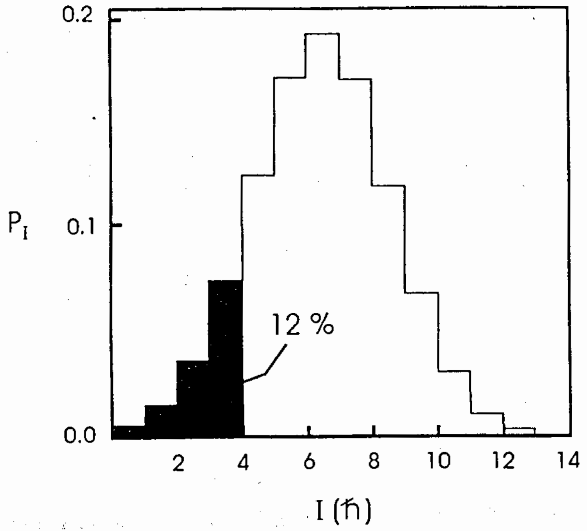

Fig. 5. Normalized spin distribution showing the calculated probability P$_I$ that the product of the ^{180m}Ta (n, n$'$) reaction retains angular momentum I ħ.

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Разрядка изомера ^{180m}Ta, индуцированная быстрыми нейтронами

Методом активации зарегистрирована разрядка изомера ^{180m}Ta ($I, K^{\pi} = 9,9^{-}$) с переходом на основное 1^{+} состояние под действием быстрых нейтронов. Фольги из естественного Ta вместе с мониторной мишенью из ^{232}Th активировались нейтронным потоком, полученным на пучке электронов с энергией 7,3 МэВ с использованием специальных конвертеров и экранов. В γ-спектре активированного Ta наблюдена $K_{\alpha 2}$ линия X-лучей Hf, принадлежащая активности ^{180g}Ta ($T_{1/2} = 8,15$ часа). Сравнивается выход трех реакций: ^{232}Th (n,f), ^{181}Ta (n,γ) ^{182}Ta и ^{180m}Ta (n,n') ^{180g}Ta при измерении в идентичных условиях. В результате определена вероятность разрядки ^{180m}Ta после рассеяния «МэВного» нейтрона. Она оказалась достаточно велика $\approx 0,4$. Установлено почти полное K-смешивание при энергии возбуждения выше энергии связи нейтрона.

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Fast Neutron Induced Depopulation of the ^{180m}Ta Isomer

The fast neutron induced depopulation of the ^{180m}Ta ($I, K^{\pi} = 9,9^{-}$) isomer to the 1^{+} ground state has been detected by activation. The natural Ta foils together with the ^{232}Th monitoring target were activated using the neutron flux produced by the 7.3 MeV electron beam at a special choice of the converters and shields. The $K_{\alpha 2}$ X-ray line (54.6 keV) of Hf has been successfully observed in the γ-ray spectrum of activated Ta. It is attributed to the activity of ^{180g}Ta ($T_{1/2} = 8.15$ h). The yields of three reactions: ^{232}Th (n,f), ^{181}Ta (n,γ) ^{182}Ta, and ^{180m}Ta (n,n') ^{180g}Ta, have been measured at the same conditions and compared. Finally, the mean probability of the ^{180m}Ta depopulation after the MeV-neutron scattering is deduced to be as high as $\approx 0,4$. Almost complete K-mixing above the neutron binding energy is established.

The investigation has been performed at the Flerov Laboratory of Nuclear Reactions, JINR.

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