Fast neutron induced depopulation of the $^{180}Ta^{m}$ isomer

S. A. Karamian,[1] C. B. Collins,[2] J. J. Carroll,[3] J. Adam,[1] A. G. Belov,[1] and V. I. Stegailov[1]
[1]*Joint Institute for Nuclear Research, 141980 Dubna, Russia*
[2]*Center for Quantum Electronics, University of Texas at Dallas, Richardson, Texas 75080*
[3]*Center for Photon Induced Processes, Department of Physics and Astronomy, Youngstown State University, Youngstown, Ohio 44555*
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Fast neutron-induced depopulation of the $^{180}Ta^{m}$ $(I,\ K^{\pi}=9,9^{-})$ isomer to the 1^{+} ground state was detected by activation techniques. Natural Ta foils together with a ^{232}Th monitoring target were activated using the neutron flux produced from a 7.3 MeV electron beam by a special choice of converters and shields. The $K_{\alpha 2}$ x-ray line (54.6 keV) of Hf was successfully observed in the γ-ray spectrum from activated Ta and attributed to the decay of $^{180}Ta^{g}(T_{1/2}=8.15\ \text{h})$. The yields of three reactions $^{232}Th(n,f)$, $^{181}Ta(n,\gamma)^{182}Ta$, and $^{180}Ta^{m}(n,n')^{180}Ta^{g}$, were measured in the same conditions and compared. Finally, the mean probability for $^{180}Ta^{m}$ depopulation after MeV neutron scattering was deduced to approach about 0.4. This establishes almost complete K mixing above the neutron binding energy. [S0556-2813(99)02602-3]

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I. INTRODUCTION

The probability of induced depopulation of isomeric states is a significant problem in the physics of nuclear reactions, as well as being important for applications in astrophysics and induced γ emission. The $^{180}Ta^{m}$ isomer survives in nature due to its half-life of $T_{1/2}\geqslant 10^{15}$ years, but its natural abundance is low, 1.2×10^{-4}, due to restrictions on its 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. These high I and K values apply strong limitations on transitions from the isomer (spontaneous and induced) in the form of selection rules and few levels with appropriate I and K quantum numbers are available in ^{180}Ta at modest E^{*}. It is worthwhile to recall that the angular momentum projection, K onto the symmetry axis is typically a well-conserved quantum number in deformed nuclei at low E^{*}.

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

In the original series of experiments [1,2], a large yield of $^{180}Ta^{g}$ activation was observed at bremsstrahlung end-point energies in the range $E_e=(2.5–5.0)$ MeV. The absolute probability of depopulation was deduced recently [4] after an experiment performed at $E_e=(5.4–7.6)$ MeV in which the yield was normalized to the well-known $^{232}Th(\gamma,f)$ reaction. The low-energy range (1.0–2.5) MeV was covered during the past year in a high-sensitivity experiment [5] performed at Stuttgart using an 150 mg, 5% enriched $^{180}Ta^{m}$ target.

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

In such a context, the development of experiments on induced isomer depopulation may be useful to obtain a phenomenology of the K-mixing amplitude dependence on the reaction, nucleus species, and excitation energy. Inelastic neutron scattering presents a new possibility for this work which was not previously studied. Slow neutrons are obviously inefficient because of the angular momentum restrictions, $\Delta I=8$, for $^{180}Ta^{m}$ depopulation. The residual energy after an (n,n') reaction ought to be at least higher than the rotational energy in order to populate the GSB at $I\geqslant(7–8)$. In the scattering of (2–3) MeV neutrons a rather low probability for depopulation is expected, comparing the residual excitation after (n,n') reactions with the photon energy in Ref. [4]. However, one has to bear in mind that inelastic scattering via compound nucleus (CN) formation is the most important mechanism for neutron scattering on heavy nuclei. Thus, CN excitation may manifest itself in intensive K mixing. Then, the residual excitation plays a passive role, and the comparison with (γ,γ') reaction results at the same excitation of the ^{180}Ta nucleus may be invalid. This situation can be clarified experimentally, as discussed below.

II. EXPERIMENTAL

An experiment to search for the $^{180}Ta^{m}(n,n')^{180}Ta^{g}$ reaction has been performed using the activation technique with a natural Ta target. MeV neutrons were produced from the

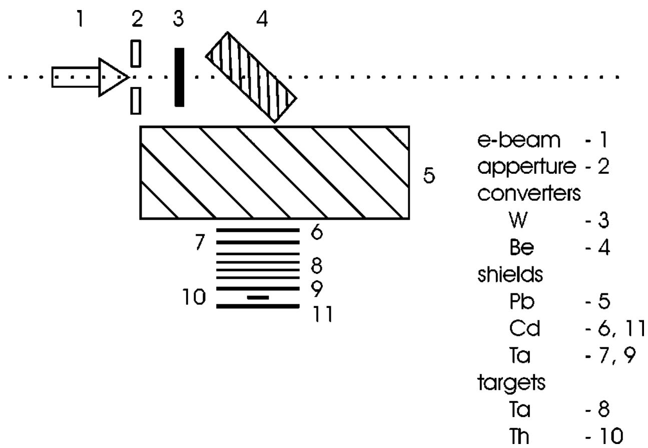

FIG. 1. Scheme of the experiment to study activation of $^{180}Ta^{g}$ by MeV neutrons.

^{9}Be (γ,n) reaction at the Dubna MT-25 microtron. The 7.3 MeV electron beam was first transformed into bremsstrahlung and then into a neutron flux by W and Be converters, respectively. The double conversion of the 20 μA electron beam produced more than 2×10^{8} neutrons/s into 4π. The activated natural Ta foils together with a ^{232}Th target used as a monitor of the neutron flux were placed behind a lead brick at a distance of about 60 mm from the Be converter at 90° from the initial electron beam direction. Thick Cd and Ta envelopes were used as shields to suppress the thermal and resonance neutron fluxes. The scheme of the irradiation is shown in Fig. 1. The choice of this geometry was necessary to avoid strong activation of Ta by slow neutrons as well as to make negligible the photon induced reaction yield. The cross-section for neutron capture by ^{181}Ta is very high at low energies, and the daughter radionuclide, ^{182}Ta $(T_{1/2}=115\ d)$, creates an intense background in induced γ-activity measurements (see below). Slow neutrons are present in the accelerator chamber, but the Cd shield completely absorbs the thermal component of the spectrum and 2 mm natural Ta plates act as resonant filters to suppress ^{182}Ta activation in the thin foils. The photon flux in the location of the activated target is decreased by many orders of magnitude due to the converter-target distance, an exposure angle far from the forward-peaked bremsstrahlung and absorption in the shields. The yield of the $^{180}Ta^{m}(\gamma,\gamma')$ reaction was measured in experiment [4] by placing a Ta target just behind the W converter. From those results, it was straightforward to calculate the reaction yield in the geometry of the present experiment, and it was found to be negligible in comparison with the expected (n,n') reaction yield. Similarly, a negligible ratio for (γ,f) compared with (n,f) reaction yields was found for the ^{232}Th target. The $^{181}Ta(\gamma,n)^{180}Ta^{g}$ reaction is completely nonexistent, since $E_e=7.3$ MeV is below the reaction threshold.

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 $^{9}Be(\gamma,n)$ reaction

$$N_n(E_n)=cN_\gamma(E_n+1.67,\ E_e)\sigma_{\gamma,n}(E_n+1.67)E_n^{-0.14}. \tag{1}$$

The reaction threshold is 1.67 MeV and the resonance behavior of the $\sigma_{\gamma,n}(E_\gamma)$ function is known from Refs. [12,13]. The last term in Eq. (1) is a correction taking into account spectral softening by the shields. The $N_n(E_n)$ function was calculated using a Monte Carlo simulated $N_\gamma(E_\gamma)$ spectrum at the end-point energy $E_e=7.3$ MeV, which corresponds to $E_n^{\max}\approx5.6$ MeV. The $\sigma_{\gamma,n}$ and neutron spectrum $N_n(E_n)$ functions are shown in Fig. 2(a). One can see the complicated form of the spectrum including several maxima. The tendency to regularly increase the neutron flux toward low E_n values is due to the bremsstrahlung spectral distribution N_γ. With these bremsstrahlung and neutron spectra, only inelastic scattering and neutron capture are opened reaction channels. Thus, no intense background activities could be produced in Ta after the irradiation other than the ^{182}Ta radionuclide.

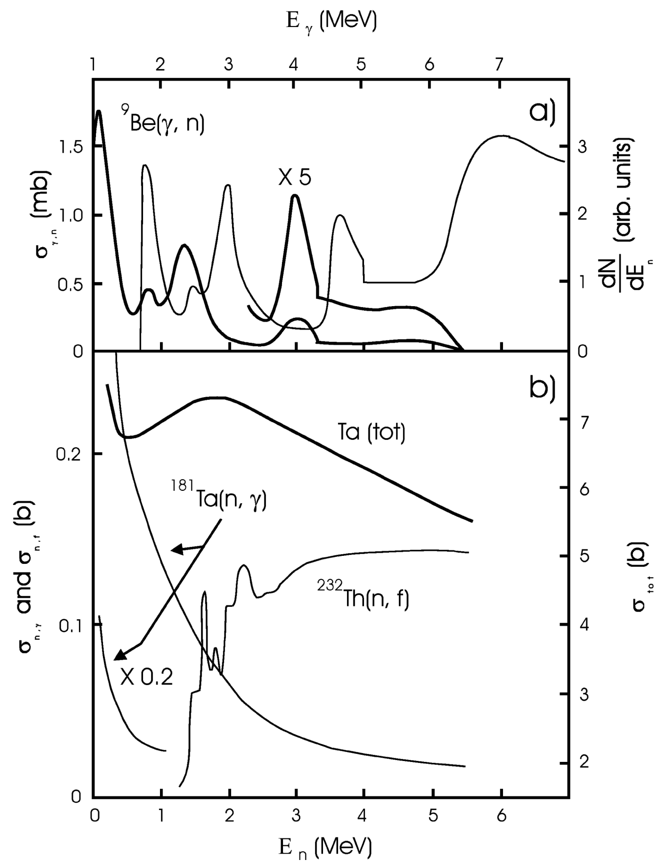

FIG. 2. (a) Cross section for the $^{9}Be(\gamma,n)$ reaction as a function of E_γ [12,13], plotted by the left-hand and top axes, and the spectral density of neutrons produced in this reaction by bremsstrahlung with $E_e=7.3$ MeV, heavy curves plotted by the right-hand and lower axes. (b) Energy dependence of the cross sections for the neutron-induced reactions $^{232}Th(n,f)$ and $^{181}Ta(n,\gamma)$, plotted by the left-hand axis, and the total reaction cross-section for ^{181}Ta, heavy curve plotted by the right-hand axis.

The 10 g $^{\mathrm{nat}}$Ta target (stack of foils) was exposed to the neutron flux for a duration of 6 h in the above-described conditions. This target contains a total of about 1 mg of $^{180}Ta^{m}$. The individual Ta foils, of 50 μm thickness, were distributed after irradiation around an HPGe detector for γ spectroscopy of the induced activity. The energy resolution of the detector was about 0.7 keV near $E_\gamma=60$ keV. Self-absorption of x rays in each foil did not exceed 10%, and the corresponding correction was introduced. The activity of $^{180}Ta^{g}(T_{1/2}=8.15\ \mathrm{h})$ was searched for in Hf K x-ray lines,

TABLE I. x- and γ-ray lines of activity detected from Ta target foils following neutron irradiation.

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

most intense in the ground-state decay. As clearly shown in Fig. 3, a weak $K_{\alpha 2}$ Hf line (54.6 keV) was detected despite the presence of more intense radiation from ^{182}Ta. This line disappeared after a cooling period of 13 h, confirming its identification with the decay of the 8 h lived ^{180}Ta^g nuclide. By subtracting the spectrum of Fig. 3(b) from that in Fig. 3(a), one can also identify the $K_{\beta 1}$ Hf line (63.2 keV) but with poor statistics. As expected, ^{182}Ta was produced in a large amount due to 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 natural radioactivity lines, were observed in the measured spectra. Unexpectedly, Ta K x rays were also very intense 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 expectations, and their affiliation is explained in Table I which indicates that some of these lines are not resolved in Fig. 3. These appear as one peak, for instance a $K_{\alpha 1}$ line from one element is typically combined with the $K_{\alpha 2}$ line of the next element.

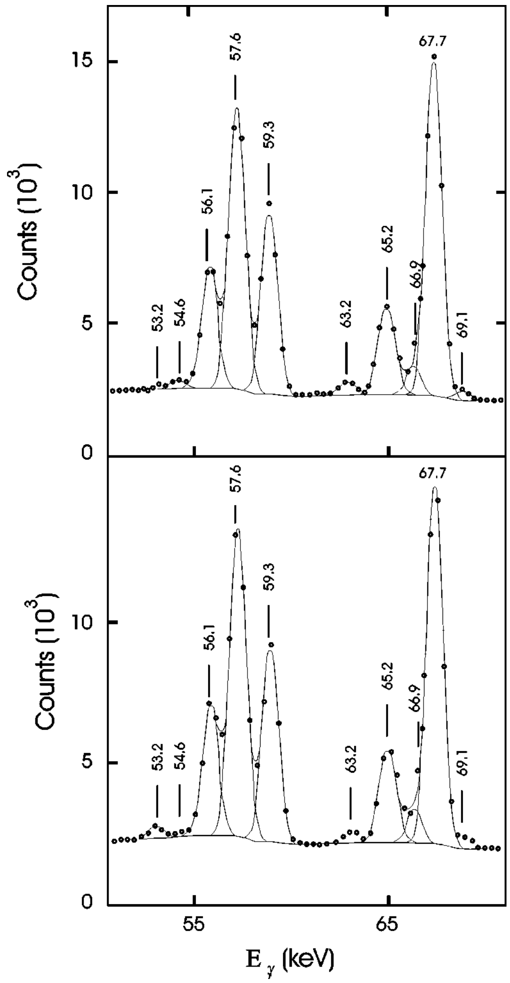

FIG. 3. γ-ray spectra from the Ta target after activation by fast neutrons measured after ''cooling'' durations of 2 h (a) and 13 h (b). The measurement time was about 11 h in both cases.

III. RESULTS AND DISCUSSION

As shown in Fig. 3, the $K_{\alpha 2}$ line of Hf was observed successfully, and the statistical inaccuracy of its area was about 15%. This allowed the determination of the integral yield of the ^{180}Ta$^m(n,n')^{180}$Ta^g reaction and a comparison with the yields of the ^{181}Ta$(n,\gamma)^{182}$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 by detecting the radioactive fragments, such as ^{91}Sr,^{92}Sr,^{97}Zr,^{133}I,^{142}La, etc. The results are given in Table II.

The experimentally determined yields were simulated using the calculated neutron spectrum, Eq. (1), and the excitation function of the reaction.

The $\sigma(E_n)$ functions for the ^{232}Th(n,f) and ^{181}Ta(n,γ) reactions are shown in Fig. 2(b) in accordance with the standard values [14]. The simulated ratio between the yields for

TABLE II. Yields of the fast neutron induced reactions in Ta target foils and the Th calibration target.

Reaction	B_n (MeV)	Mean E_n (MeV)	CN Mean E^* (MeV)	Normalized yield Expt.	Normalized yield Simulated
^{232}Th (n,f)	4.79	2.7	7.5	1	1
^{181}Ta$(n,\gamma)^{182}$Ta	6.06	0.42	6.5	6.2 ± 0.2	6.18
^{180}Ta$^m(n,n')^{180}$Ta^g	7.65	2.3	9.9	13.4 ± 3.8	13.4[a]

[a]If $R=\sigma_g/(\sigma_g+\sigma_m)=0.403$ is used.

these two reactions perfectly reproduced the measured value, the comparison being given in Table II.

For the $^{180}\mathrm{Ta}^m(n,n')^{180}\mathrm{Ta}^g$ reaction neither its cross section nor energy dependence are known. The following threshold behavior for the excitation function was introduced in order to estimate the probability of the depopulation based on the measured yield:

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

where the threshold energy is expressed as

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

The value 0.7 MeV corresponds to the rotational energy for $I=8$ in the ^{180}Ta residual nucleus. The mean kinetic energy of the scattered neutron, $\varepsilon_{\mathrm{kin}}(n')$ was calculated assuming the mechanism of statistical evaporation from the CN. Above the threshold the cross-section is defined by the neutron absorption cross section (equal to $0.5\sigma_{\mathrm{tot}}$ in the black nucleus model) and by the probability for decay of the CN to the ground state: $\mathrm{R}=\sigma_g/(\sigma_g+\sigma_m)$. The total reaction cross section σ_{tot} is shown in Fig. 2(b) in accordance with the data of Ref. [14] for the ^{181}Ta nucleus. It is impossible to expect any significant difference in the total cross section between $^{180}\mathrm{Ta}^m$ and ^{181}Ta nuclei, since the geometric parameters define σ_{tot} for MeV neutrons. The mean CN excitation is defined by the mean projectile energy $\overline{E_n}$, calculated as follows:

$$\overline{E_n}=\int_0^{E_n^{\max}} E_n\sigma(E_n)N_n(E_n)dE_n \Big/ \int_0^{E_n^{\max}} \sigma(E_n)N_n(E_n)dE_n. \tag{4}$$

Numerical values are given in Table II.

Equations (1)–(3) were used to simulate the yield of the $^{180}\mathrm{Ta}^m(n,n')^{180}\mathrm{Ta}^g$ 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. This means that the probability is as high as 40% for isomer depopulation to the g.s. after (n,n') reactions. Such a high probability was not expected taking into account the results of Ref. [4] for a (γ,γ') reaction at 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 lost immediately after neutron absorption at sufficiently high excitation of the compound nucleus (above 9 MeV). The subsequent emission of a neutron and γ rays is governed by the standard statistical laws. Accordingly, the residual excitation will definitely exceed the rotational energy [Eq. (3)], and all transitions should satisfy the selection rules on spin and other quantum numbers. In photon scattering the nucleus does not pass through a highly excited compound nucleus stage and this influences the probability of $^{180}\mathrm{Ta}^m$ depopulation, giving different values for the two reactions at the same residual excitation.

Accepting this explanation, the value of $\mathrm{R}=\sigma_g/(\sigma_g+\sigma_m)=0.4$ determined from the (n,n') reaction experiment is placed in Fig. 4 at the position of mean CN excitation, not mean excitation of the residual nucleus for comparison with

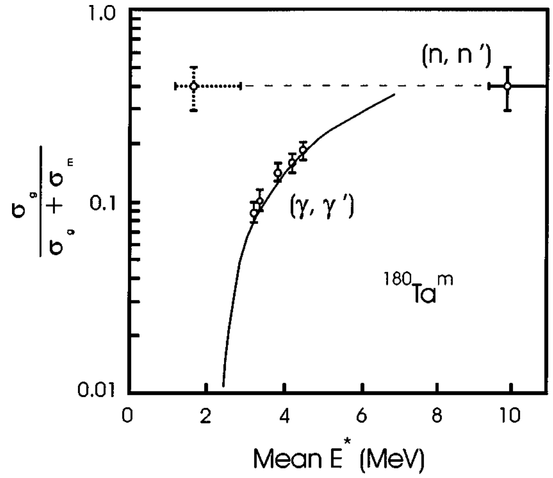

FIG. 4. Probability for $^{180}\mathrm{Ta}^m$ isomer depopulation measured in (γ,γ') and (n,n') reactions as a function of the mean compound nucleus excitation energy. The dashed point indicates the misleading placement of the (n,n') datum if it were located by the residual excitation energy.

(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 accuracy of the measured depopulation probability R includes statistical error (15%) and 10% error due to the normalization and simulation procedures. The horizontal error 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 a quasimonochromatic neutron spectrum. Until then it is difficult to comment upon the energy dependence of the cross section, although its threshold behavior seems to be specific to the $^{180}\mathrm{Ta}^m(n,n')^{180}\mathrm{Ta}^g$ reaction. In any event, almost complete K mixing at $E^*\geqslant B_n$ in deformed nuclei is confirmed as clearly seen from Fig. 4.

Stretched γ cascades with regularly decreasing I value are typical in heavy deformed nuclei, being cascades via rotational states within bands built on the levels of different nature. The K-quantum number is conserved within the individual bands. Conversely, stretched cascades with regularly decreasing K value are unknown. An assumption of such a peculiar behavior for ^{180}Ta is not confirmed by the results of (γ,γ') reaction studies. Indeed, for the latter reaction the isomer depopulation probability remains modest up to an excitation energy that is higher than the mean residual excitation in the (n,n') reaction. So, for an understanding of the present results it is necessary to use the idea of K mixing as discussed above. The only alternative would be 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 $^{180}\mathrm{Ta}^m(n,n')$ reaction product was calculated for input(output) kinetic energies of the neutron of 3.0(1.5) MeV and at

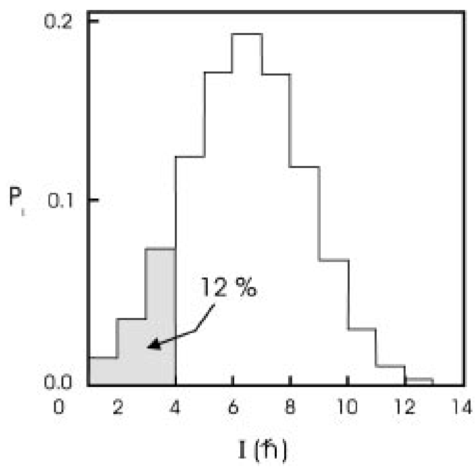

FIG. 5. Normalized spin distribution showing the calculated probablity P_I that the product of the ^{180}Ta$^m(n,n')$ reaction retains angular momentum $I\hbar$. Only 12% corresponds to the low spin part with $I \leqslant 4$.

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% of the total isomer depletion probability corresponds to the low-spin part of the distribution with $I \leqslant 4$. This is not sufficient to explain the 40% probability for the reaction branch ending up in the 1^+ g.s. in ^{180}Ta. The value $\Delta K = 3$ can be released with a reasonable probability in a cascade of 2–3 statistical γ quanta. However, $\Delta K = 4$ 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 γ's cannot account for a large part of the release of $\Delta K = 8$ in the $(n,n'\gamma)$ reaction. The only explanation is the existence of K-mixing properties for intermediate compound-nucleus states as discussed above.

IV. CONCLUSION

The probability for ^{180}Ta^m isomer depopulation to the ground state by an inelastic neutron scattering reaction was measured for the first time. The results show definitively that the specific K configuration of the isomer does not survive after 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.

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