Isomeric to ground state ratio in the $^{180}Ta^{m}(\gamma,\gamma')^{180}Ta^{m,g}$ reaction

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The yield Y of the $^{180}Ta^{m}(\gamma,\gamma')^{180}Ta^{g}$ ($T_{1/2}$=8.15 h) reaction has been measured as a function of the bremsstrahlung end-point energy E_e in the range of 2.5–7.6 MeV. An activation technique was used for the yield measurement which was calibrated by monitoring the $^{232}Th(\gamma,f)$ reaction yield. The $Y(E_e)$ function was numerically simulated and the probability $\sigma_g/(\sigma_g+\sigma_m)$ for the depletion of the $^{180}Ta^{m}$ isomer after γ-ray absorption was deduced. This probability is important for astrophysical questions and schemes for producing induced γ emission and perhaps a γ-ray laser. [S0556-2813(98)06104-4]

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I. INTRODUCTION

The only isomer which survives in nature is $^{180}Ta^{m}$, having a half-life $T_{1/2}\geqslant 10^{15}$ yr and a low abundance of 1.2×10^{-4}. The isomer is identified as a two-quasiparticle deformation-aligned configuration with I, $K^{\pi}=9,\ 9^{-}$ and an excitation energy of 75 keV. The depletion of $^{180}Ta^{m}$ to the ground state in (γ,γ') reactions has previously been studied in Refs. [1–3]. A quite large yield of the radioactive $^{180}Ta^{g}$ nucleus was found [2] following the irradiation of targets containing $^{180}Ta^{m}$ in natural abundance with bremsstrahlung at end-point energies $E_e\geqslant 3$ MeV, while a sharp cutoff of the yield was observed at $E_e\leqslant 2.5$ MeV. Thus, the energy of the first activation level was determined to be near 2.8 MeV and a value of integrated cross section for the reaction was established. The absolute probability for isomer depletion resulting from γ-ray absorption is a crucial parameter both in ''optical'' schemes for induced γ emission or γ-ray laser pumping, and in studies of the isomer lifetime within stellar conditions. The potential advantages of ''optical'' pumping of a γ-ray laser with a population beginning in a long-lived isomeric level have been discussed in the review article of Ref. [4]. Despite these studies, the absolute probability for isomer depletion has not been directly measured. The present work represents a first attempt to accomplish this objective.

II. THEORETICAL BACKGROUND

An isomeric state usually has a single particle configuration that is significantly different from that of the ground state and is often characterized by high values of the spin and spin projection quantum numbers I and K. The decay of such an isomer to the ground-state band is retarded by the electromagnetic selection rules for the change of I, K, and parity. Therefore, the excitation (or depletion) of an isomeric state in a nuclear reaction can be expected to be reduced by the spin difference in accordance with the statistical model and by structure selectivity (ΔK hindrance). It has been assumed theoretically and recently experimentally confirmed [5] that the selection rule on the K quantum number does not survive at high excitation energies $E^{*}>B_n$ (B_n is the neutron binding energy). Thus one can conclude the existence of complete K mixing at such high energies. However, below B_n a different situation should exist and additional experiments are necessary to clarify the problem in detail. A model for isomer population or depopulation by (γ,γ') reactions via absorption by special activation levels was introduced many years ago. Due to the large values of integrated cross section found for those reactions, those activation levels were assumed to serve as intermediate states for structure-type transformations which provide significant K mixing even at modest excitation energies. However, the specific properties of such levels are not yet clear.

The results of previous experiments [1–3] on $^{180}Ta^{m}$ depopulation were analyzed according to the assumption that only individual activation levels are manifested within the limits of experimental uncertainty and integrated cross sections for the respective resonances were deduced on this basis. Such an approach was appropriate at the modest excitation energies of those studies since activation levels were resolvable even using continuous bremsstrahlung spectra. However, for excitation energies $E^{*}\geqslant 4$ MeV many levels may contribute to the observed yield, and the analysis must be modified since individual transitions may not be resolved experimentally. In the present work the cross section for isomer depletion is taken to be a product of two factors:

$$\sigma(E_\gamma)=\sigma_{\mathrm{GDR}}(E_\gamma)\frac{\sigma_g}{\sigma_g+\sigma_m}(E_\gamma), \tag{1}$$

where $\sigma_{\mathrm{GDR}}(E_\gamma)$ is the cross section of the giant electric dipole resonance for γ-ray absorption by a nucleus at the energy E_γ and $\sigma_g/(\sigma_g+\sigma_m)$ is the probability for decay of the compound nucleus to the ground state. Both functions in Eq. (1) are considered to be separate functions of the continuous variable E_γ. The quantity $\sigma_g/(\sigma_g+\sigma_m)$ is in effect a branching ratio, but reflects all possible branches, direct or via cascade, which reach the ground state after the excitation of an activation level. It contains the contributions of all K-mixing levels within a selected E_γ interval. For a significant probability to exist for the excitation step from isomer to activation level by photon absorption, an electric dipole

transition is most likely. The form of Eq. (1) reflects the expectation that at energies below B_n, the strength in the tail of the giant electric dipole resonance bounds the cross section for individual $E1$ transitions.

The yield of the bremsstrahlung-induced reaction is then expressed as

$$Y(E_e)=c\int_0^{E_e}\sigma(E_\gamma)N_\gamma(E_\gamma)dE_\gamma, \tag{2}$$

where c characterizes the experimental irradiation geometry and other efficiency factors, and $N_\gamma=d^2W/dedE_\gamma$ is the spectral density of the bremsstrahlung radiation normalized to one electron in the beam. The data of Ref. [3] was consistent with isolated activation levels at energies $E_\gamma\leqslant 4$ MeV and so the analysis was based on that structure. At the higher energies of this study, γ-ray absorption yield is averaged over many levels and contains the product of GDR cross section and spectral density of radiation in accordance with the standard approximation. The $\sigma_{\rm GDR}(E_\gamma)$, by definition a strength function, represents the absorption cross section per E_γ interval and is therefore independent of the number of levels in the interval. Thus the new experimental results are analyzed in a framework that is explicitly self-consistent to provide the absolute probability of $^{180}\mathrm{Ta}^m$ depletion by the (γ,γ') reaction.

Equations (1) and (2) are used to simulate the energy dependence seen in experimentally measured yields. This formulation is typical for experiments utilizing a continuous energy spectrum for the incident radiation. Likewise it is consistent with methods employed in the analysis of questions in stellar nucleosynthesis and scenarios for ''optical'' pumping of a γ-ray laser.

III. EXPERIMENTAL DETAIL

The $^{180}\mathrm{Ta}^m(\gamma,\gamma')^{180}\mathrm{Ta}^g$ reaction yield, $Y(E_e)$ was measured in this work using bremsstrahlung at end-point energies of E_e = 5.4, 6.2, 7.0, and 7.6 MeV. These results are combined with the data of Ref. [2] in order to plot the yield function over a wider range of end-point energies from 2.5 to 7.6 MeV. An important improvement in experimental technique, available due to the higher end-point energies, was the calibration of the $^{180}\mathrm{Ta}^g$ yield by comparison with the yield from a monitoring reaction, $^{232}\mathrm{Th}(\gamma,f)$detected in the same experimental conditions. This permitted a determination of absolute values of cross section for the (γ,γ') reaction. A comparison with the total absorption cross section enabled the reaction probability $\sigma_g/(\sigma_g+\sigma_m)$ to be deduced.

The measurements were obtained using the electron beam of the MT-25 microtron at FLNR, Dubna. Targets of $^{\rm nat}$Ta and ThO_2 were placed behind the electron beam converter (2.5 mm tungsten) for activation by bremsstrahlung. A stack of 50 μm-thickness Ta foils with a total mass of 0.7 g was used as the $^{\rm nat}$Ta target. This stack was disassembled for measurements of x- and γ-ray spectra to decrease the thickness of the source, and a correction factor was introduced to account for the remaining self-absorption of low-energy photons within the foils. The spectra were measured by an HPGe detector with an energy resolution of 0.95 keV as determined by lines of ^{57}Co. By taking into account decay and efficiency

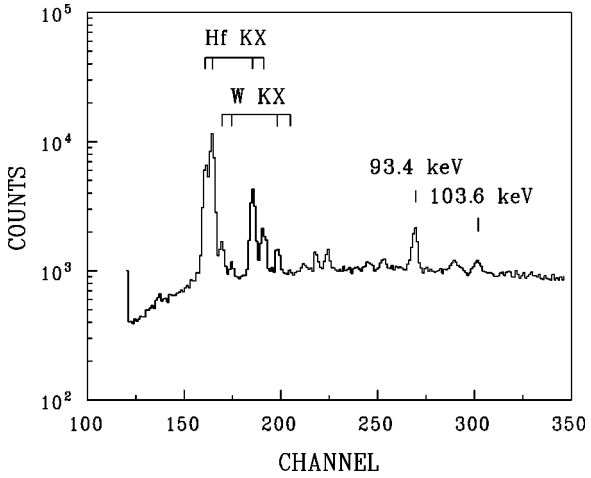

FIG. 1. Gamma spectrum obtained from a natural Ta target following its activation by bremsstrahlung having an end-point energy of E_e=6.2 MeV. Marked peaks are indicative of daughters resulting from the decay of $^{180}\mathrm{Ta}^g$.

factors, the γ-line intensities were evaluated to determine the number of radioactive atoms $N_{\rm at}^{\rm prod.}$ produced in the irradiations, and the yield of the reaction normalized to one target nucleus and to one electron of the microtron beam:

$$Y=\frac{N_{\rm at}^{\rm prod.}}{N_{\rm at}^{\rm tar.}N_e}, \tag{3}$$

where $N_{\rm at}^{\rm tar.}$ is the number of target atoms and N_e is the time-integrated number of beam electrons during the irradiation. The γ-line abundances and other properties of the radioactive nuclides were taken from Ref. [6].

A rich γ spectrum of fission fragments was recorded for the ThO_2 target. The excitation function of the $^{232}\mathrm{Th}(\gamma,f)$ reaction has been measured with high accuracy [7,8] and the fission fragment mass distribution has been studied in Ref. [9]. These data are sufficient to calibrate the yield when fission products are detected in specific experimental conditions.

X-ray and γ-ray lines belonging to $^{180}\mathrm{Ta}^g$ decay ($T_{1/2}$=8.15 h, lines of Hf KX, W KX, and E_γ=93.4 and 103.6 keV) were detected in spectra obtained after the Ta target activation and the line intensities were measured with good statistical accuracy. A typical measured spectrum is shown in Fig. 1. In addition to the intense $^{180}\mathrm{Ta}^g$ lines, background lines are observed in the figure, particularly Pb KX rays which are present from the detector shielding. The natural radioactivity line at 92.5 keV was subtracted from the area of the 93.4 keV γ peak from $^{180}\mathrm{Ta}^g$. The decay curves of the latter line and of the Hf $K_{\alpha_1}+K_{\alpha_2}$ lines are shown in Fig. 2 in comparison with calculated curves based on the known half-life of $^{180}\mathrm{Ta}^g$, $T_{1/2}$ = 8.15 h. There was excellent agreement between the measured data and the decay calculations. This and the correct ratio of the line areas insured that pure $^{180}\mathrm{Ta}^g$ was detected without any background contribution.

In general, when $^{\rm nat}$Ta targets are used, reactions on the abundant isotope ^{181}Ta can create an intense background in

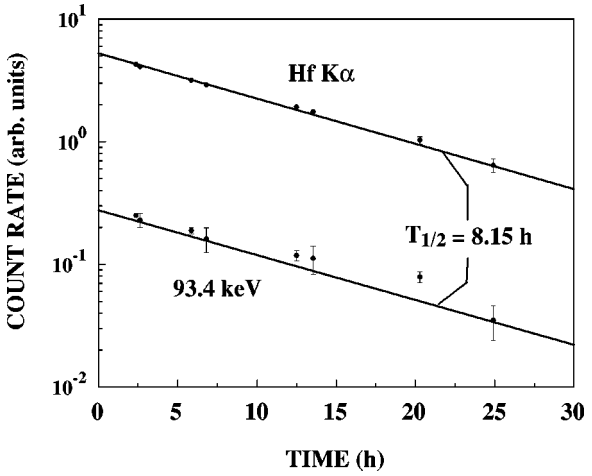

FIG. 2. Time spectrum showing the decay of the Hf $K_{\alpha_1}+K_{\alpha_2}$ and 93.4 keV γ lines which signify production and transmutation of $^{180}\mathrm{Ta}^{g}$ following activation of a target foil.

spectroscopic measurements. However, for these experiments the contribution of the $^{181}\mathrm{Ta}(\gamma,n)\,^{180}\mathrm{Ta}^{g}$ reaction was excluded at $E_e \leqslant 7.6$ MeV, recalling that the neutron binding energy $B_n=7.58$ MeV for the ^{181}Ta nucleus. The detected yield of $^{180}\mathrm{Ta}^{g}$ was wholly due to the $^{180}\mathrm{Ta}^{m}(\gamma,\gamma')$ reaction despite the very low $^{180}\mathrm{Ta}^{m}$ concentration of 1.2×10^{-4} in the target. For the cross section analysis, it is important to mention that (γ,γ') processes are the dominant mode of compound nucleus deexcitation below the threshold for neutron emission. The value $B_n=6.6$ MeV was deduced for $^{180}\mathrm{Ta}^{m}$ from nuclear mass tables, but due to the requirement of spin conservation it is necessary to add the yrast energy of about 0.5 MeV. Therefore, the $^{180}\mathrm{Ta}^{m}(\gamma,n)$ reaction can be competitive with the (γ,γ') processes only at $E_\gamma \geqslant 7.3$ MeV, allowing 0.2 MeV for the neutron kinetic energy. Quite a few γ quanta with such E_γ are created at $E_e=7.6$ MeV. For these reasons the maximum end-point energy in the present experiment was chosen to be $E_e=7.6$ MeV. The minimum end point $E_e=5.4$ MeV was necessary because of accelerator limitations. The minimum present end point of 5.4 MeV is close to the maximum E_e for the experiments of Ref. [2] and so the yields measured there could be linked with the present results. Thus the yield function $Y(E_e)$ was determined over the wide range $E_e=2.5$–7.6 MeV as shown in Fig. 3. One can see a regular growth of the yield at $E_e \geqslant 3.5$ MeV in accordance with expectations, and physically significant conclusions can be deduced after analysis of these data.

IV. RESULTS

For the yield simulation, bremsstrahlung spectra normalized to one beam electron were calculated using the EGS4 Monte Carlo code in a configuration developed at the University of Texas at Dallas. Examples of the spectra corresponding to end-point energy values of $E_e=5.5$, 6.5, and 7.5 MeV are given in Fig. 4. In accordance with the close irradiation geometry, the spectra were calculated as integrals over the forward hemisphere. The yield of $^{232}\mathrm{Th}(\gamma,f)$ was evaluated [see Eq. (2)] based on the calculated N_γ function and measured $\sigma_f(E_\gamma)$ [7,8] for this reaction. By comparison

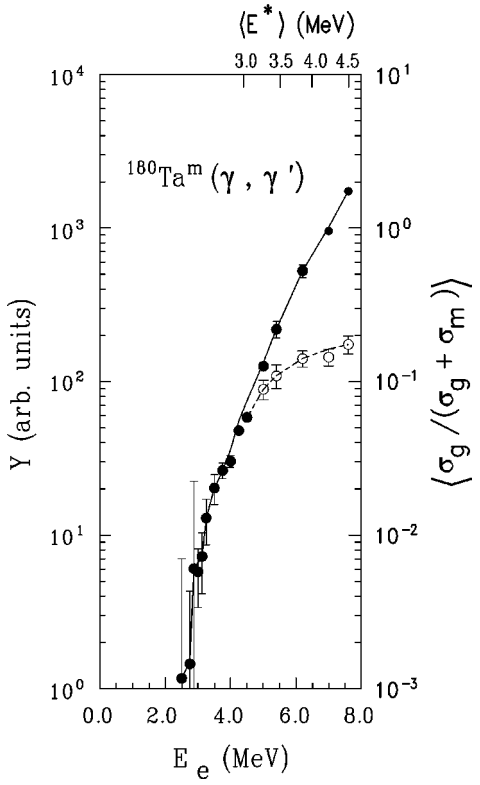

FIG. 3. Measured yields (closed circles plotted with the left-hand axis) for the reaction $^{180}\mathrm{Ta}^{m}(\gamma,\gamma')\,^{180}\mathrm{Ta}^{g}$ as a function of bremsstrahlung end-point energy. Mean excitation energies for various end points are shown by the top axis. Open circles plotted by the right-hand axis show the mean probability for isomer depletion, that is, assuming a constant value over each bremsstrahlung continuum at the corresponding mean excitation energy. Curves are drawn to guide the eye to the experimental points.

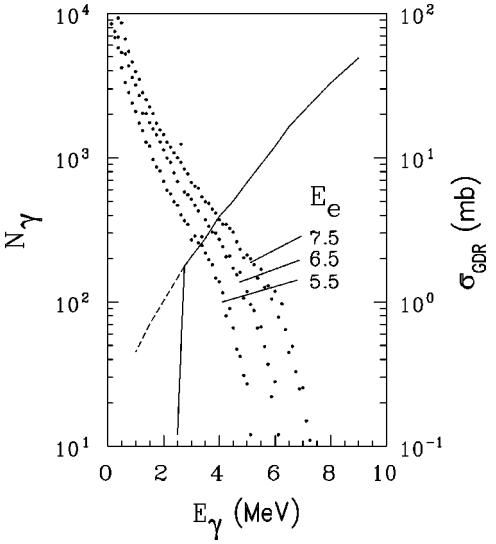

FIG. 4. Typical bremsstrahlung spectra calculated with the EGS4 code plotted with the left-hand axis. The right-hand axis plots the function used to represent the photon absorption cross section in the low-energy tail of the giant electric dipole resonance. The solid curve for this cross section includes a cutoff employed for the $R=$const approach, while the actual values below 2.8 MeV (dashed section) were used for the $R=R(E_\gamma)$ analysis.

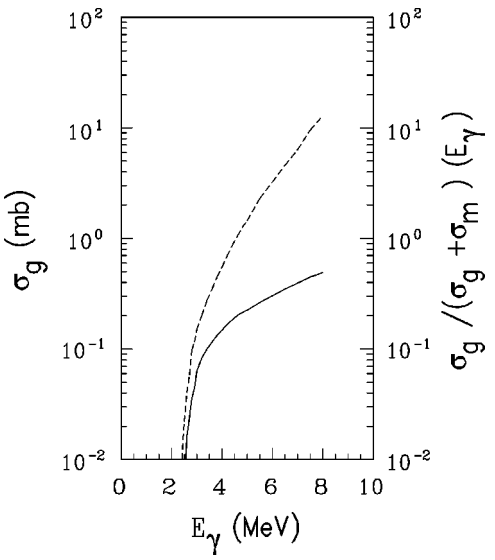

FIG. 5. Calculated absolute cross section σ_g (dashed curve plotted by the left-hand axis) and isomer depopulation probability $\sigma_g/(\sigma_g+\sigma_m)$ (solid curve plotted by the right-hand axis) for the $^{180}\mathrm{Ta}^m(\gamma,\gamma')^{180}\mathrm{Ta}^g$ reaction as a function of the excitation energy E_γ of an activation level.

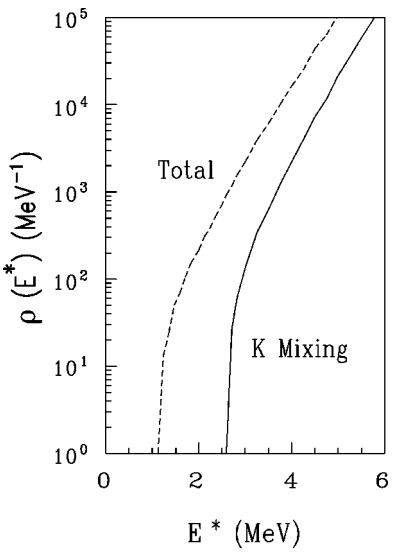

FIG. 6. Calculated level densities for all states, ρ (dashed curve), and states having K-mixing properties for the depopulation of $^{180}\mathrm{Ta}^m$, $\rho_I(E^*)$ (solid curve).

with the experimentally-determined yield value [Eq. (3)], the experimental parameter c was found and used in the analysis of the data for the $^{180}\mathrm{Ta}^m(\gamma,\gamma')$ reaction.

The γ-ray absorption yield was calculated for the $^{180}\mathrm{Ta}^m$ nucleus with the normalization provided by the coefficient c for the actual experimental geometry. After comparison with the measured yield, the ratio $R=\sigma_g/(\sigma_g+\sigma_m)$ was deduced immediately under the assumption of $R=$const. Taking R to be independent of E_γ at a specific E_e value is equivalent to averaging this ratio over the E_γ range, weighted by the function $\sigma_{\mathrm{GDR}}(E_\gamma)\cdot N_\gamma$ [see Eqs. (1) and (2)]. The mean value of the excitation energy E^* can also be calculated. The $\sigma_{\mathrm{GDR}}(E_\gamma)$ function was taken from the data of Ref. [10] for the ^{181}Ta nucleus, as shown in Fig. 4. However, as is evident from the figure, an artificial cutoff was introduced at $E_\gamma \leqslant 2.8$ MeV to avoid a spuriously large contribution from intense, low-energy portions of the bremsstrahlung continua. At present, there are no measurements to support the existence of activation levels below that energy for $^{180}\mathrm{Ta}^m$ depopulation as discussed in Ref. [2].

The measured $Y(E_e)$ function and deduced mean $R=\sigma_g/(\sigma_g+\sigma_m)$ values are shown in Fig. 3. The ratio R increases as E_e grows and reaches a magnitude of about 0.18. The error is on the level of 15% and arises from the normalization and other data processing procedures. The statistical accuracy is much better as seen from the error bars of the yield measurements. Finally, the isomeric-to-ground state ratio $\sigma_m/\sigma_g=4.5$ was found for the reaction $^{180}\mathrm{Ta}^m(\gamma,\gamma')^{180}\mathrm{Ta}^{m,g}$, deviating significantly from the results of Ref. [3]. The ratio σ_m/σ_g is large, thus incomplete K mixing takes place at this modest mean excitation energy. This is in contrast to the essentially complete K mixing at higher energies deduced from the systematics of Refs. [5,11]. One can expect that the ratio R increases at larger energies to approach 0.5 near $\langle E^*\rangle \approx 7$ MeV in accordance with the systematics. However, it is difficult to attain such $\langle E^*\rangle$ values using bremsstrahlung irradiations without employing an endpoint energy greater than 7.6 MeV, unsuitable as explained above. Thus experiments with a quasimonochromatic γ-ray source are necessary to pursue this question further. At present, this type of source is not widely available at the required intensity.

One can try to overcome these difficulties by employing a more sophisticated analytical approach to simulate the yield function with Eqs. (1) and (2) using $R=R(E_\gamma)$. In this analysis, the truncation of the σ_{GDR} function below 2.8 MeV was not applied. Of course, this cutoff must appear naturally (if it exists) in the behavior of $R(E_\gamma)$ obtained from the yield simulation procedure. The values for $R(\langle E^*\rangle)$ shown in Fig. 3 were used as a first iteration. The method was to determine the $R(E_\gamma)$ function over a wider energy range by sequential iteration using a mathematical parametrization of this function and the minimum χ^2 criterion for reproducing the measured $Y(E_e)$ values. The result is shown in Fig. 5. The absolute value of R at $E_\gamma=7$ MeV is about 0.4, not far from the systematics prediction of Refs. [5,11].

Figure 5 can be considered as the first direct description of K-mixing behavior over a wide range of excitation energies in the following manner. In γ-ray capture by the isomeric nucleus, a specific set of compound states with known quantum numbers I^π is excited due to spin and parity conservation. Among these states one can distinguish K-conserving and K-mixing levels. The K-conserving levels inherit from the isomer ($K=K_i$) a similar $K=K_i\pm 1$ and so those states can decay only into bands with values of K very similar to that of the isomer. It is reasonable to assume that 70% of the decay strength eventually feeds back into the initial isomeric state. In contrast, K-mixing levels should not be influenced at all by the K value of the isomer and can decay into any band with any K value, although the spin and parity selection rules must still be satisfied. The deformed nucleus normally has many bands feeding the ground state and much fewer bands linked with the band built on the initial isomeric state. Hence, it is clear that a K-mixing level will decay to the ground state band with about 90% branch-

ing with only a small portion of the total intensity feeding back into the isomer. In such a schematic model, it is easy to understand that the ratio $R=\sigma_g/(\sigma_g+\sigma_m)$ approximates the ratio of the density of K-mixing states to the total level density. The total density $\rho_I(E^*)$ of levels with specific spin I can be calculated using the standard statistical model, for instance, the often used formulation of Ref. [12]. The product

$$R\cdot\rho_I(E^*)=\rho_I^{\mathrm{mix}}(E^*) \tag{4}$$

then gives the K-mixing level density $\rho_I^{\mathrm{mix}}(E^*)$ for states of the same spin I. The calculated level densities (both K-mixing and total) for the case of $A=180$ and $I=9$ are presented in Fig. 6. It is interesting to see that near an excitation energy of $E^*=2.5$ MeV, the K-mixing level density is quite low, about one level per MeV. That is in accordance with previous measurements [2] that the first activation level for $^{180}\mathrm{Ta}^m(\gamma,\gamma')$ depopulation is near 2.8 MeV. In addition, one can see in Fig. 6 a definitive onset of the K-mixing level density function. That means there exists a threshold for the K-mixing mode excitation, which may be explained in the style of Ref. [11] by considering the potential energy surface parametrized by γ deformation. Triaxially deformed states can be excited above this barrier which are not K conserving because of the broken axial symmetry. A microscopic explanation of this mechanism remains for future studies.

V. SUMMARY

The normalized yield of the $^{180}\mathrm{Ta}^m(\gamma,\gamma')^{180}\mathrm{Ta}^g$ reaction has been measured over the range of bremsstrahlung endpoint energies of $E_e=2.5$–7.6 MeV. The absolute probability for isomer depopulation was deduced from simulations of the measured yield values. Incomplete K mixing was concluded at these excitation energies and the energy dependence of the density of K-mixing levels was evaluated.

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