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Resonant excitation of the reaction $^{180}Ta^m(\gamma,\gamma')\,^{180}Ta$

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Irradiation with a superconducting linear accelerator of Ta has provided data for the characterization of the reaction $^{180}Ta^m(\gamma,\gamma')\,^{180}Ta$. The depopulation of the isomer $^{180}Ta^m$ via an intermediate state or narrow band of states near 2.8 MeV has been found with an integrated cross section of 1.2×10^{-25} cm^2 keV. This large value exceeds, by nearly an order of magnitude, known cross sections for (γ,γ') reactions producing isomers of other species. Another intermediate state or narrow band is also indicated by the data at an energy 0.6 MeV higher.

The isotope $^{180}Ta^m$ is nature's rarest stable nuclide[1] being only 0.012% of all tantalum and the only naturally occurring isomer.[2] However, the importance of $^{180}Ta^m$ lies not in its rarity but in its abundance. The nucleus ^{180}Ta sits somewhat aside the main path of the *s* process[3,4] for cosmic nucleosynthesis and the survival of any amount into current times raises some difficult questions resulting from the presence of the isomer. The ground state of ^{180}Ta has a half-life of only 8.1 h while the isomer has an energy of 75.3 keV and a half-life[2] of 1.2×10^{15} y. The branching of nucleosynthesis to the ground and metastable states is obviously important, but even after creation populations may continue to transfer between these levels by photoexcitation, altering the effective half-life of the nucleus and the understanding of its present abundance. Either this isomer must have been singularly stable against photonuclear deexcitation, $^{180}Ta^m(\gamma,\gamma')\,^{180}Ta$ at the time of creation,[5,6] or the corresponding temperatures must have been too low to produce photons capable of pumping such a reaction.[7]

These latter concerns have been aggravated by the most recent experiments.[8] Not only does photonuclear deexcitation of $^{180}Ta^m$ occur, the integrated cross section reported for the process is of unprecedented size for a (γ,γ') reaction connecting ground state and isomer. However, that result was obtained by irradiating an enriched sample of $^{180}Ta^m$ with the bremsstrahlung continuum from a 6 MeV linac and so the energies of the particular photons pumping the reaction could not be determined. Reported here is the measurement of an excitation function between 2 and 5 MeV and the discovery of a very large integrated cross section in excess of 10^{-25} cm^2 keV for the deexcitation of $^{180}Ta^m$ by 2.8 MeV photons.

The energy-level diagram of ^{180}Ta and its daughters is shown in Fig. 1, together with a representation of some steps in the excitation and detection of the $^{180}Ta^m(\gamma,\gamma')\,^{180}Ta$ reaction. The principal means for the detection of the ^{180}Ta ground state lies in observing the $K\alpha$ lines of its daughter, ^{180}Hf following decay by electron capture. The efficiency for the emission of $K\alpha$ photons relative to the number of ^{180}Ta decays[9] is about 57%.

The time integrated yield of ground-state nuclei, N_f obtained by irradiating N_i isomers with a photon flux Φ_0 in photons/cm^2 delivered in a bremsstrahlung continuum of intensities up to an end point energy E_0 is,

$$N_f = N_i\Phi_0\int_0^{E_0}\sigma(E)F(E,E_0)dE\ , \tag{1a}$$

where $F(E,E_0)$ is the distribution of intensities within the bremsstrahlung spectrum normalized so that

$$\int_0^{E_0}F(E,E_0)dE = 1\ , \tag{1b}$$

and $\sigma(E)$ is the cross section for the reaction

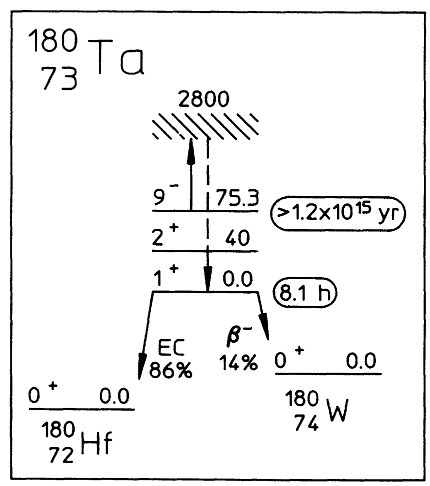

FIG. 1. Schematic energy-level diagram of ^{180}Ta and its daughters. Half-lives are shown in ovals for the ground and isomeric levels. Energies are in keV. The initial transition of the (γ,γ') reaction is shown by the arrow pointing upward to the intermediate state represented by the hatched line. Cascade through the levels of ^{180}Ta is not known, but leads finally to the ground state, which decays as indicated by the diagonal downward arrows.

$^{180}Ta^m(\gamma,\gamma)^{180}Ta$ as a function of the photon energy E.

All (γ,γ') reactions below particle threshold energies excite nuclear bound states. Therefore production or depletion of isomers by these reactions proceeds through resonant excitation of intermediate states, each with rather narrow widths. One such channel is shown in Fig. 1 at an excitation energy of $E_j = 2.8$ MeV.

The odd-odd nucleus ^{180}Ta, having a particularly high density of states, could have intermediate states lying at high energies with separations comparable to their widths, in which case the integral of Eq. (1a) could then be simplified no further. However, at lower energies where a discrete number of intermediate states contribute, the spectral intensity $F(E,E_0)$ will vary little over the narrow range of energies for which $\sigma(E)$ is nonzero around each of the E_j. Then the ground-state yield, expressed as the normalized activation per unit photon flux $A_f(E_0)$ produced with bremsstrahlung having an end point of E_0 can be written from Eq. (1a) as,

$$A_f(E_0) \equiv \frac{N_f}{N_i \Phi_0} = \sum_j (\sigma\Gamma)_{fj} F(E_j,E_0) \,. \tag{2a}$$

In this expression $(\sigma\Gamma)_{fj}$ is the integrated cross section for the production of ground-state N_f as a result of the excitation of the intermediate state E_j with bremsstrahlung described by the spectral function $F(E,E_0)$, so that

$$(\sigma\Gamma)_{fj} = \int_{E_j-\Delta}^{E_j+\Delta} \sigma(E)\, dE \,, \tag{2b}$$

where Δ is an energy small compared to the spacing between intermediate states and large in comparison to their widths. Levels of this type are sometimes called gateways or doorways. The integrated cross sections for such levels can be evaluated with the residue, $R_M(E_0)$ obtained by subtracting the contributions to A_f from excitations through M intermediate states,

$$R_M(E_0) = A_f(E_0) - \sum_{E_j=E_1}^{E_M} (\sigma\Gamma)_{fj} F(E_j,E_0) \,. \tag{3}$$

A change of the end point energy E_0 of the bremstrahlung, as well as altering Φ_0, modulates the spectral intensity function $F(E_j,E_0)$ at all of the important energies for resonant excitation, E_j. The largest effect occurs when E_0 is increased from a value just below some intermediate state at $E_j = E_k$ to one exceeding it so that $F(E_k,E_0)$ varies from zero to some finite value.

Early work[10] on (γ,γ') reactions showed that a plot of activation, $A_f(E_0)$ as a function of bremsstrahlung end point, E_0 displayed very pronounced activation edges at the energies, E_j corresponding to the resonant excitation of new intermediate states. Unfortunately, such an excitation function was not reported previously[8] for the reaction $^{180}Ta^m(\gamma,\gamma')^{180}Ta$, so the question was unresolved as to whether or not the extraordinary size found for the integrated cross section $(\sigma\Gamma)$ was the result of many smaller $(\sigma\Gamma)_{fj}$ summing to a large value as suggested by Eq. (2a). At the time of that experiment there was no source of bremsstrahlung with a variable end point and enough intensity to provide significant excitation of samples available in such minute amounts as $^{180}Ta^m$.

In the work reported here bremsstrahlung was obtained from a Ta converter foil irradiated by the electron beam from the injector of the 130 MeV superconducting Darmstadt linear accelerator (*S*-DALINAC) at the Technische Hochschule Darmstadt.[11] The electrons were accelerated in three superconducting cavities in which the continuous wave rf amplitudes were varied to change the electron energy (here in the range from 2 to 5 MeV). The diameter of the electron beam was about 2 mm and this and other beam parameters were monitored and kept constant. Uncertainty in the end point was less than 50 keV.

The numbers of final-state nuclei N_f were obtained in these experiments by detecting signature photons with a Ge(Li) spectrometer. Counts in the appropriate channels were corrected for the finite durations of both irradiation and counting, for the absolute counting efficiency of the spectrometer, for the emission intensity relative to the parent, and for the opacity of the experimental sample to the escape of signature photons. The latter factor was calculated with a Monte Carlo code.

Samples used in the experiments for the deexcitation of $^{180}Ta^m$ were disks 3.8 cm in diameter and 127 μm thick. The material was 99.95% pure tantalum and contained $^{180}Ta^m$ in its natural abundance. Irradiations were made for a nominal 4 h period at a beam current of 20 μA. The actual charge passed to the bremsstrahlung converter was determined by digitizing the current and numerically integrating it during the irradiation interval. Planchettes containing nominal amounts of 2 g of SrF_2 in natural isotopic abundances were concurrently exposed in contact with the Ta foils for calibration purposes. Fourteen different end points of the bremsstrahlung were arranged to span the interval from 2 to 5 MeV.

The evaluation of $A_f(E_0)$ in Eq. (2a) requires knowledge of the particular spectral intensity functions, $F(E,E_0)$, together with the photon flux, Φ_0, incident on each sample position. These were calculated for the different end point energies, E_0 with the established EGS4 coupled electron-photon transport code developed at SLAC.[12] Verification of the calculated values of flux could only be obtained by the reaction $^{87}Sr(\gamma,\gamma')^{87}Sr^m$. This has been distinguished in the literature[10] by the comprehensive report of its excitation energies E_j and integrated cross sections $(\sigma\Gamma)_{mj}$ for production of metastable states at energies below 3 MeV and therefore serves as a benchmark for the analysis of these experiments.

The dependence of the values of $A_f(E_0)$ for $^{87}Sr^m$ upon the bremsstrahlung end point was determined from the measurements of its 388.4 keV decay signature fluorescence.[9] The dominant[10] activation edge near 2.67 MeV was well reproduced. The residue $R_3(E_0)$ was computed for the three intermediate state locations indicated in Ref. 10, leaving their integrated cross sections variable. The values of integrated cross sections best describing the data are shown in Table I. Below 4 MeV, the results of this work are in remarkable agreement with the previous measurements and thus verify the calculations of $F(E,E_0)$.

The increase of residues above 4 MeV suggested the importance of another intermediate state. The final entry in Table I records the integrated cross section found to be sufficient to describe observations of $A_f(E_0)$ over the en-

TABLE I. Values of integrated cross section $(\sigma\Gamma)_{fj}$ for the reaction $^{87}Sr(\gamma,\gamma')^{87}Sr^{m}$ through gateway states indicated by the measured excitation function. The gateway excitation energies E_j for levels in Ref. 10 are given at the previously determined locations. The energy of the new state indicated by this work is given at the centroid of the appropriate spectral bin.

Energy (MeV)	$\sigma\Gamma(\times 10^{-29}$ cm^{2}keV) Ref. 10	This work
1.22	8.5+4−3	8.5±2
1.88	16+8−5	16±4
2.67	380+200−100	430±50
4.3±0.1	· · ·	1500±300

ergy range up to 5 MeV.

The thermal and fast neutron fluxes in the irradiation environment were measured by standard techniques[13] and were found to give negligible contributions to the excitation function for ^{87}Sr.

The depopulation of $^{180}Ta^{m}$ was examined by observation of the Hf K x-ray signatures from ^{180}Ta ground-state decay. Background subtracted spectra of the data corrected for counting times shown in Fig. 2 clearly display the lowest energy activation edge for the reaction $^{180}Ta^{m}(\gamma,\gamma')^{180}Ta$.

Figure 3(a) plots the values of $A_f(E_0)$ obtained in this work for the deexcitation reaction $^{180}Ta^{m}(\gamma,\gamma')^{180}Ta$ and shows the dependence associated[14] with the increase of E_0 above an intermediate state. The measured excitation strictly excludes broad band photoabsorption such as due to the density of states or the tail of the giant dipole resonance, which must contribute at least a constant integrated cross section in each spectral bin above 2.8 MeV. The properties of the lowest energy intermediate state are the most important to the question of the survival of $^{180}Ta^{m}$ during cosmic nucleosynthesis. A strong level or narrow band of states near 2.8 MeV can be rather well determined by the data.[15]

Figure 3(b) shows the residue computed from Eq. (3) with $M=2$ to remove the contributions from the two lowest intermediate states or narrow bands observed in this work. The values of integrated cross section we found for deexcitation through these levels are shown in Table II.

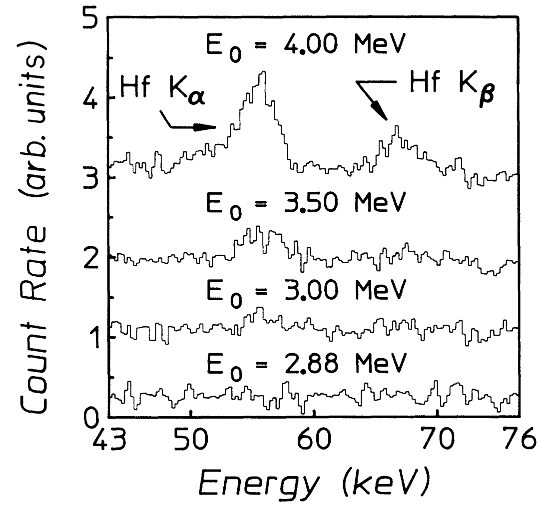

FIG. 2. Spectra of fluorescent photons from ^{180}Ta decay after irradiation with bremsstrahlung having the end points shown. The counting rate from an unexposed sample has been subtracted.

It is important to consider some likely contaminants to these data. Neutron capture is not possible since the analogous parent ^{179}Ta does not exist naturally. Also, photodissociation of ^{181}Ta can produce some $^{180}Ta^{g}$, but the threshold for this process is 7.576 MeV. The remaining reaction to consider is $^{181}Ta(n,\gamma)^{182}Ta$, leading to ^{182}W with a characteristic x-ray which could contribute to the broad structure attributed to Hf $K\alpha$ in Fig. 2. However, the 115 d half-life of ^{182}Ta made possible delayed measurements uncontaminated by $^{180}Ta^{g}$. These showed, even at the highest end point of 5 MeV, null results and indicated that no contamination could have occurred from this process.

The results of this work are in close agreement with the previous measurements[8] indicating an integrated cross section for $^{180}Ta^{m}(\gamma,\gamma')^{180}Ta$ of extraordinary size. However, it is now possible to report the excitation energy for the lowest strong level for the deexcitation of $^{180}Ta^{m}$ at 2.8 MeV. The corresponding value of the integrated

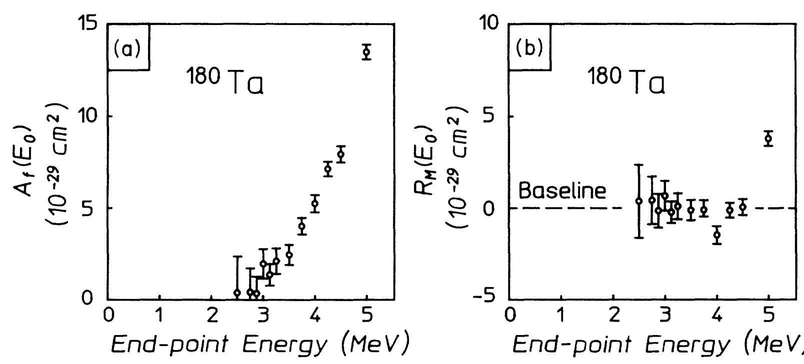

FIG. 3. (a) Linear plot of activation $A_f(E_0)$ for ^{180}Ta as a function of the bremsstrahlung end point. (b) Residue computed from Eq. (3) after removing the contributions from the two lowest energy gateways for deexcitation using parameters recorded in Table II.

TABLE II. Values of integrated cross section $(\sigma\Gamma)_{fj}$ for the reaction $^{180}Ta^m(\gamma,\gamma')^{180}Ta$ through gateway states indicated by the excitation function of Fig. 3(a). The gateway excitation energies E_j for these levels are given at the centroid of the appropriate spectral bins.

Energy (MeV)	$\sigma\Gamma(\times 10^{-29}$ cm^{2}keV)
2.8 ± 0.1	$12\,000 \pm 2000$
3.6 ± 0.1	$35\,000 \pm 5000$

cross section is 1.2×10^{-25} cm^{2}keV and is the largest ever reported for a (γ,γ') reaction connecting a ground state and an isomer at energies below the threshold for the evaporation of neutrons. Notwithstanding the singular magnitude of this cross section, the state's energy for photodeexcitation of $^{180}Ta^m$ is just high enough[7] to insure the survival of this nucleus in the stellar environment and current models of cosmic nucleosynthesis are sustained.

The nuclear structure of these intermediate states for this well deformed, odd-odd nucleus also presents an interesting problem. The extraordinary $\Delta K = 8$ needed to reach the ^{180}Ta ground state implies considerable K mixing of these levels. A possible scheme which explains the large upward transition probability and the sudden onset of the depopulation would be as follows: At low energies, numerous K-allowed transitions can be constructed from the Nilsson model states of the unpaired proton and neutron. Here, K mixing is small and the states will entirely decay back to the isomer. In the simple Nilsson model a few levels with $\Delta K = 0,1$ with respect to the isomer are possible at energies near 3 MeV which can be excited by enhanced $E1$ transitions. Due to the high level density of ^{180}Ta, large K mixing would then result in depopulation of the isomer to the ground state. A detailed analysis of this process is currently underway.[16]

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[15]The data below 3 MeV allow the existence of weaker intermediate states at lower energies. Reference 6 provided a null result for ground-state production at 1.3 MeV and the present work indicates that a level near 1.4 MeV would only have an integrated cross section of 500×10^{-29} cm^{2}keV.
[16]P. Vogel (private communication).