Photoactivation of $^{180}Ta^{m}$ and Its Implications for the Nucleosynthesis of Nature's Rarest Naturally Occurring Isotope

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The depopulation of the quasistable isomer in ^{180}Ta with $J^{\pi} = 9^{-}$ at $E_x = 75$ keV by resonant photoabsorption was investigated with intense bremsstrahlung. The results indicate a dramatic acceleration of the isomer decay to the short-lived $J^{\pi} = 1^{+}$ ground state under stellar s-process conditions. The consequences for a possible nucleosynthesis of nature's rarest isotope ^{180}Ta within the s process are discussed.

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The exotic odd-odd nucleus ^{180}Ta carries a dual distinction. It is nature's rarest isotope with a relative abundance of only 0.012% of the rarest element tantalum (0.02 on a scale where silicon is normalized to 10^6). This tiny fraction owes its survival to a long-lived isomer ($t_{1/2} > 1.2 \times 10^{15}$ yr [1]) at an excitation energy $E_x = 75$ keV with $J^{\pi} = 9^{-}$ which is stabilized by the large spin difference to the $J^{\pi} = 1^{+}$ ground state. The latter decays with a half-life of 8.15 h. Therefore, all ^{180}Ta is found in the isomeric state, and it is the only naturally occurring isomer.

In spite of considerable experimental and theoretical efforts the nucleosynthesis of $^{180}Ta^{m}$ still represents an unsolved puzzle. Surprisingly, the problem lies not in the rarity, but rather in identifying a mechanism producing sufficient amounts of this isotope [2]. On one hand, ^{180}Ta lies aside the main path of the s process which proceeds through the chain of stable Hf isotopes directly to ^{181}Ta. On the other hand, it is shielded against the r process by its stable isobar ^{180}Hf. A variety of possible production mechanisms has been considered including explosive scenarios such as the p process [3] or neutrino nucleosynthesis [4] during type II supernovae, spallation reactions in the interstellar medium [5], and photodisintegration [6]. Apart from these more speculative scenarios, a closer look reveals two possible explanations based on more complex paths within the s process. Beer and Ward [7] suggested a β decay branch from the $J^{\pi} = 8^{-}$ isomer in ^{180}Hf which would preferentially populate the 9^{-} state in ^{180}Ta. However, the experimentally observed branching ratio seems to exclude a dominance of this reaction path [8]. Yokoi and Takahashi noticed [9] that low-lying states of ^{179}Hf become unstable against β decay under typical s-process conditions, enabling a production of $^{180}Ta^{m}$ via neutron capture on the unstable ($t_{1/2} = 665$ d) nucleus ^{179}Ta. A quantitative analysis of this important possibility has to await a successful measurement of the neutron capture cross section [10,11]. At present, a synthesis of $^{180}Ta^{m}$ in the s process is neither proven nor disproven [12]. However, recent stellar model calculations find an s-process contribution of about 50% [13].

The typical s-process temperatures of a few 10^8 K pose a further complication. The accompanying photon bath might result in a thermal coupling between isomer and ground state (gs) via resonant excitation of higher lying levels acting as intermediate states (IS), as depicted schematically in Fig. 1. For excitation energies of IS below 1 MeV the coupling could be efficient enough to destroy any ^{180}Ta synthesized in the s process. The search for astrophysically relevant IS has attracted considerable interest lately. Depopulation of the isomer was observed with large yields in photoexcitation experiments using bremsstrahlung [12,14,15], but the IS energies (2.8 and 3.6 MeV [15]) were too high to play a role here. Attempts to excite states at lower energies ($E_x < 1.3$ MeV) with intense ^{60}Co sources showed no signal [12,16].

Recently, Coulomb excitation was suggested as an alternative probe emphasizing the role of different multipoles ($E2, E3$) compared to resonant photoabsorption where dipole transitions dominate [17]. A thick-target Coulomb activation experiment using heavy ion (^{32,36}S) beams showed yields consistent with an IS at energies below 1 MeV. However, subsequent studies with thin targets enriched in ^{180}Ta suggest [18] a background reaction as source of the signal observed in [17]. In Coulomb activation with light ions, which does not suffer from comparable background problems, the experimental uncertainties of the deduced IS energies are too large to conclude on their impact on s-process production [11]. In-beam Coulomb excitation experiments have been

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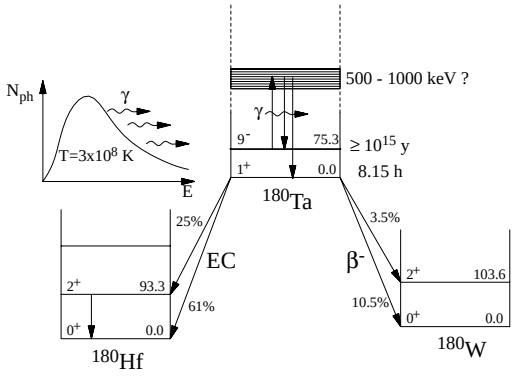

FIG. 1. Schematic energy-level diagram of ^{180}Ta and its daughters illustrating the possibility for thermally enhanced decay of ^{180}Ta^m in the stellar environment of the *s* process. The inset shows the photon density at *s* process temperature.

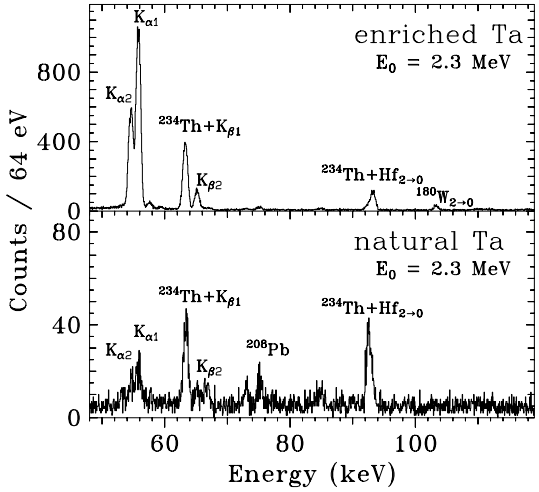

FIG. 2. Low-energy γ-ray and x-ray spectra of Ta samples from photoactivation at $E_0 = 2.3$ MeV using enriched $^{180}Ta_2O_5$ (top) and natural metal foils (bottom). Note the difference in the ordinate scales illustrating the enormous improvement in sensitivity.

reported [19,20], but their analysis is not yet completed. Several γ-spectroscopic studies searched for states in ^{180}Ta which simultaneously decay to gs and isomer, but none could be identified [21,22].

It is the purpose of this Letter to report a new photoactivation experiment with considerable improvements, e.g., a factor of 10 gain in photon fluxes, an optimized detection system, and the use of a target made out of the world's resources of highly enriched ^{180}Ta. We are able to demonstrate thermal coupling between isomer and gs for photon energies as low as about 1 MeV, resulting in a strong modification of the effective lifetime of ^{180}Ta at *s*-process temperatures.

The photoactivation experiments were performed at the bremsstrahlung irradiation facility newly installed at the Stuttgart Dynamitron accelerator [23]. dc electron currents up to 450 μA could be used on the water-cooled bremsstrahlung production target. To achieve the highest possible photon flux the distance between radiator target and Ta sample was only about 9 cm. In total 150 mg of Ta_2O_5 enriched to 5.45% in ^{180}Ta^m were available, corresponding to 6.7 mg of ^{180}Ta^m. Alternating with this enriched target, 0.2 mm thick metal foils of natural Ta were irradiated at higher bombarding energies when experimental sensitivity allowed. In addition, a sample consisting of 150 mg natural Ta oxide, prepared in exactly the same way as the enriched target, was activated at some energies to verify the enrichment factor. Typical irradiation times were about two half-lives (12–16 h).

Different methods were applied to monitor the photon beam characteristics including measurements of the electron current and of the total photon flux with an ionization chamber, mixed target (Al/Cu/LiF) nuclear resonance fluorescence investigations [24], and the well-known [25] photoactivation of the ^{115}In isomer ($E_x = 336$ keV, $t_{1/2} = 4.5$ h). For this purpose indium foils were simultaneously irradiated directly behind the Ta samples. These photon flux measurements were found to agree well with Monte Carlo simulations using the codes GEANT and EGS [25]. The bremsstrahlung end-point energies were determined from the rotating voltmeter of the Dynamitron and checked by simultaneous photon scattering experiments using lead, ^{27}Al, and ^{13}C targets, respectively.

After the irradiations, characteristic x-rays and γ-radiation from the activated samples were measured off-line by two well-shielded, high-resolution, low-energy photon (LEP) Ge detectors. Experimental details will be given in a forthcoming article. Typical spectra depicted in Fig. 2 show characteristic x-rays and nuclear γ-transitions specific for the decay of the ^{180}Ta ground state. They are dominated by the well-resolved K_{α_1} and K_{α_2} lines of Hf, the corresponding K_{β_1} and K_{β_2} peaks being also visible. Unfortunately, there is an accidental overlap of a background line following β decay of ^{234}Th with the $2^+ \rightarrow 0^+$ γ-transition in the residual nucleus ^{180}Hf. In the spectra of the enriched sample even the $2^+ \rightarrow 0^+$ transition in ^{180}W is observed, in spite of its weak feeding in the β^- decay of the ^{180}Ta (see Fig. 1). Another proof for observing the ground state decay is a precise confirmation of the lifetime in a time differential measurement covering about eight half-lives ($t_{1/2}^{\text{exp}} = 8.15 \pm 0.03$ h).

Irradiations were performed for bremsstrahlung end-point energies $E_0 = 0.8$–3.1 MeV (see Fig. 3). The expected improvement in sensitivity between the enriched Ta_2O_5 sample, metallic Ta foils, and the natural Ta_2O_5 sample was experimentally verified. The yields obtained from the different target types agree well within the statistical uncertainties. Depopulation of the isomer was observed down to $E_0 \simeq 1$ MeV. The availability of the enriched Ta sample, together with the new setup, has improved the sensitivity of the present study by a factor of about 4000 compared to previous experiments [15].

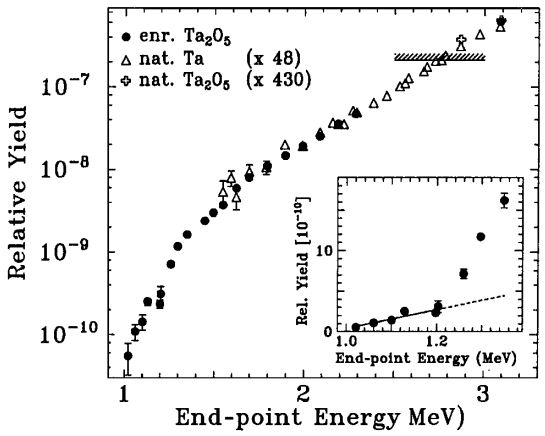

FIG. 3. Comparison of normalized Hf x-ray yields obtained with the enriched Ta_2O_5 sample (full dots), metallic Ta foils (open triangles), and the natural Ta_2O_5 sample (crosses); all values are normalized to the accumulated charge on the radiator target and to equal irradiation and measuring times. The hatched area represents the sensitivity limit of Collins *et al.* [15]. Inset: Expanded view of the experimental threshold region. The solid line represents the yields from an IS with $E_\gamma = 1.01$ MeV and $I_s = 5.7$ eV fm^2.

The most important parameter with respect to a possible destruction of ^{180}Ta^m under stellar conditions is the excitation energy of the lowest IS. The data do not permit a distinction whether the onset of gs population at $E_\gamma \simeq 1$ MeV corresponds to the lowest IS or just reflects the sensitivity limit. Assuming the former case as a conservative limit, one can extract the excitation energy and integrated cross section I_s which is related to the partial decay width $\Gamma_{\rm iso}$ of the IS by $I_s = (\pi\hbar c/E_\gamma)^2 g b_0 \Gamma_{\rm iso}$. Here, $g = (2J_f + 1)/(2J_i + 1)$ denotes a statistical factor of the initial and final state spins J_i and J_f, respectively, and b_0 stands for the summed branching ratio to decay cascades feeding the ground state. All data points below $E_0 = 1.2$ MeV, where the next higher IS is located, were included in the analysis (see Fig. 3 inset). One finds a transition energy $E_\gamma = 1.010(10)$ MeV [corresponding to $E_x(\mathrm{IS}) = 1.085$ MeV] and $I_s = (5.7 \pm 0.4^{\rm stat} \pm 1.1^{\rm syst})$ eV fm^2. The quoted systematic error is estimated from the uncertainties of the absolute photon flux calculated by GEANT Monte Carlo simulations.

The effect of an IS with these properties on the survival of ^{180}Ta^m in the presence of a stellar photon bath is calculated by solving the coupled differential equations for the three-level system isomer $\leftrightarrow$ IS $\leftrightarrow$ gs including the time needed to reach thermal equilibrium (see, e.g., [10]). For thermal energies above $kT \simeq 14$ keV a dramatic reduction of the effective half-life is observed (thick line in Fig. 4). This strong enhancement of the stellar decay rate of ^{180}Ta^m has an immediate impact on its possible *s*-process origin since it falls right into the temperature regime of He burning zones in red giant stars where the *s* process proceeds. Essentially two approaches can be invoked for describing the *s* process between Zr and Pb and the concomitant ^{180}Ta^m production: the canonical model [2] which assumes static neutron irradiations at constant temperature or fairly realistic models [27] of thermally pulsing stars on the asymptotic giant branch (AGB).

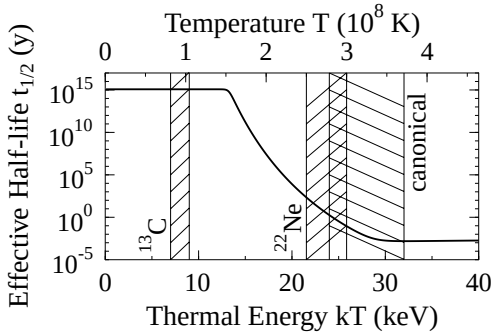

FIG. 4. Reduction of the ^{180}Ta^m half-life (thick line) in the presence of a stellar photon bath deduced from the photoactivation cross sections of the present experiment. Thermal energy ranges expected under *s*-process conditions are indicated for the canonical model [2] (\\\), and for a more realistic model [26] which distinguishes burning phases where different (α, n) reactions serve as neutron sources (///).

Temperature limits for the canonical approach have been deduced from the analysis of *s*-process branchings [28,29]. As shown in Fig. 4, they range around thermal energies of $kT = 28$ keV. At this temperature, the ^{180}Ta^m half-life being about 1.5 d implies an average survival probability reduced to less than 5% compared to the situation assumed in Ref. [12], where this enhancement was neglected. Thus, the *s*-process yield of ^{180}Ta^m predicted by the static canonical approach becomes negligibly small for realistic values of temperature and mass density.

Stellar *s*-process models for low mass $[(1.5-3)M_\odot]$ AGB stars have been considerably improved during the past decade and were shown to reproduce the *s* abundances remarkably well [26]. In these models the *s* process is determined by two neutron sources, the (α, n) reactions on ^{13}C and ^{22}Ne, which are alternatively activated during He shell burning. With respect to the production of ^{180}Ta it is important to note that the two sources operate at significantly different temperatures and time scales. About 95% of the total neutron exposure is provided by the ^{13}C(α, n) source during the low temperature phase of shell hydrogen burning, characterized by low neutron densities and thermal energies around $kT = 8$ eV (see Fig. 4). While the corresponding temperature of 10^8 K leaves the ^{180}Ta half-life unaffected, it is also too low for ^{179}Hf to become unstable. Thus, only the weak route via the ^{180}Hf isomer to ^{180}Ta remains effective under these conditions.

Significant production of ^{180}Ta is obtained, however, during the relatively short He shell flashes, which last about 100 yr and follow the much longer periods of hydrogen shell burning ($\approx$50 000 yr). The enormous

energy production during the He shell flash causes a highly convective region with an extension of about $10^{-2}M_{\odot}$ and typical turnover times of about one day [30]. Towards the end of the shell flash, the temperatures in the reaction zone near the bottom of the convective region rise to 250 to 300 million K, thus igniting the ^{22}Ne source, which then yields an intense neutron burst for a few years. Although this burst contributes only about 5% to the overall exposure, it suffices to rearrange the abundance patterns of the *s*-process branchings, which, therefore, reflect the higher temperatures during the He shell flashes.

Typically, thermal energies of $kT = 26$ keV are reached at maximum under these conditions (see Fig. 4), enough to initiate the decay of ^{179}Hf and thereby opening the additional production mechanism of ^{180}Ta via neutron captures on ^{179}Ta. The final yield from this mechanism is intimately related to the survival probability of ^{180}Ta and, hence, to the time period for which this isotope is exposed to high temperatures. Because of the steep decrease of the half-life with temperature, the survival of ^{180}Ta depends sensitively on the convection time scale: While its half-life is about one month at the temperature of the reaction zone, freshly produced ^{180}Ta is mixed to the cooler layers of the He shell flash within a few hours due to the short turnover time of about one day.

Accordingly, most of the ^{180}Ta is expected to survive this scenario. Including the temperature dependence of the ^{180}Ta half-life of Fig. 4 into the detailed stellar model calculations of Ref. [13] and using the calculated cross sections reported in Ref. [12] yields in fact the same *s*-process contribution of 50% as in the previous approach where the enhanced decay was neglected. Very likely, 50% may even be a lower bound since a recent measurement [31] indicates a much smaller ^{180}Ta neutron capture cross section, corresponding to a larger *s* abundance as compared to the assumptions made in [12].

In summary, we have demonstrated the depopulation of the long-lived isomer in ^{180}Ta to the short-lived ground state by resonant absorption of photons through a possible intermediate state with energies as low as 1.01 MeV. This leads to a dramatic reduction of the effective half-life in the photon bath accompanying the *s* process. Nevertheless, the resultant lifetime is still long compared to the short convection time of about one day predicted in recent stellar *s*-process models. If the present scenario is to explain the ^{180}Ta production freshly synthesized material would have to be rapidly mixed into cooler zones. Accordingly, ^{180}Ta is not precluded from being produced by the *s* process unless intermediate states could be identified at even lower energies. Therefore, experimental efforts to further improve the sensitivity achieved in the present work would be of high importance.

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